

# Unified approach to QED in arbitrary linear media

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We give a unified approach to macroscopic QED in arbitrary linearly responding media, based on the quite general, nonlocal form of the conductivity tensor as it can be introduced within the framework of linear response theory, and appropriately chosen sets of bosonic variables. The formalism generalizes the quantization schemes that have been developed previously for diverse classes of linear media. In particular, it turns out that the scheme developed for locally responding linear magnetodielectric media can be recovered from the general scheme as a limiting case for weakly spatially dispersive media. With regard to practical applications, we furthermore address the dielectric approximation for the conductivity tensor and the surface impedance method for the calculation of the Green tensor of the macroscopic Maxwell equations, the two central quantities of the theory.

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## I. INTRODUCTION

In both classical and quantum electrodynamics, it is often advisable to divide, at least notionally, the matter that interacts with the electromagnetic field into a part that plays the role of a passive background and a remainder, active part that needs to be considered in more detail. By means of suitable coarse-graining and averaging procedures, this leads to the well-known framework of Maxwell's phenomenological equations, where the background—the medium—is treated as a continuum and, quite frequently, by the methods of linear response theory. From this perspective, the characterization of the medium is reduced to the prescription of suitable constitutive relations, i.e., appropriate response functions or susceptibilities.

Depending on the specific kinds of media under consideration, it is under many circumstances sufficiently accurate to work with spatially local response functions, taking into account only (temporal) dispersion and absorption in accordance with causality. For conducting and semiconducting media (not to mention plasmas) as well as superconducting materials, however, the spatially local description can be inadequate due to the existence of almost freely movable charge carriers (conduction electrons, excitons, Cooper pairs) in such media. Hence, if one is not willing to restrict one's attention to a crude spatial resolution and/or specific frequency windows, spatial dispersion, i.e., the spatially non-local character of the medium response, generally cannot be disregarded for such media. Electrodynamics problems with the inclusion of spatial dispersion have been considered by various authors in different ways, both on the classical and quantum levels; for classical approaches, see, e.g., Refs. [1–7], for quantum ones see, e.g., Refs. [8, 9].

A scheme that takes spatial dispersion into account along with dispersion and absorption in sufficiently general terms can also be regarded as an important step towards a satisfactory (quantum) electrodynamics of mov-

ing media, which is very much lacking at present. The reason is that a medium, even if it can be assumed to respond spatially locally when it is at rest, will in general appear as responding non-locally when it is in motion. Given that the polarization of typical Drude–Lorentz-type dielectrics responds to the electric field with a characteristic memory time of the order of  $10^{-9} \dots 10^{-7}$  s [10], already moderate (i.e., non-relativistic) velocities may lead to the appearance of noticeable spatial non-localities. For example, sonoluminescence experiments show that the collapse of a bubble with a typical initial radius of  $10 \dots 50 \mu\text{m}$  to a final radius of around  $1 \mu\text{m}$  occurs on a time scale similar to the characteristic memory time of the response of the surrounding fluid [11].

The study of the quantized electromagnetic field in spatially non-locally responding media and the prospect of elaborating a quantum theory of light in moving media will also open up new ways of investigating quantum effects related to the recently proposed ‘optical black hole’ [12, 13]. So far, the theory has concentrated on purely geometrical optics with some progress being made towards a (scalar) wave-optical description, but a consistent linear-response approach is still lacking.

The paper is organized as follows. In Sec. II we introduce the basic concepts of field quantization in arbitrary linearly responding media, with special emphasis on spatially dispersive media. This serves as the basis for a detailed study of possible choices of appropriate dynamical variables in Sec. III. We then proceed to show in Sec. IV how previously introduced quantization schemes for diverse classes of media can be obtained as special cases from the general quantization scheme developed in Sec. II. In addition to the general formalism, some knowledge of the structure of the Green tensor for spatially dispersive media is needed when performing explicit calculations. This problem is addressed in Sec. V, where it is described (in general and by an example) how the surface impedance method may be applied in this context, on the basis of the dielectric approximation. Some

concluding remarks are given in Sec. VI.

## II. QUANTIZATION SCHEME

The effect of any linear, dispersing and absorbing medium on the electromagnetic field can be described, within the framework of linear response theory, by the relation

$$\underline{\mathbf{j}}(\mathbf{r}, \omega) = \int d^3r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{E}}(\mathbf{r}', \omega) + \underline{\mathbf{j}}_N(\mathbf{r}, \omega), \quad (1)$$

where  $\underline{\mathbf{j}}(\mathbf{r}, \omega)$  and  $\underline{\mathbf{E}}(\mathbf{r}, \omega)$ , respectively, are the (linearly responding) current density and the electric field in the frequency domain,  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is the complex conductivity tensor in the frequency domain [7, 14], and  $\underline{\mathbf{j}}_N(\mathbf{r}, \omega)$  is a Langevin noise source. According to the Onsager reciprocity theorem [7, 14], the conductivity tensor should be reciprocal,  $Q_{ij}(\mathbf{r}, \mathbf{r}', \omega) = Q_{ji}(\mathbf{r}', \mathbf{r}, \omega)$ . Except for a translationally invariant (bulk) medium, the spatial arguments  $\mathbf{r}$  and  $\mathbf{r}'$  of  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  must be kept as two separate variables in general. We assume that, for chosen  $\omega$ ,  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is the integral kernel of a reasonably well-behaved (integral) operator acting on vector functions in position space. In particular, we assume that  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  tends (sufficiently rapidly) to zero for  $|\mathbf{r} - \mathbf{r}'| \rightarrow \infty$  and has no strong (i.e., non-integrable) singularities (specifically, for  $\mathbf{r}' \rightarrow \mathbf{r}$ ). To allow for the spatially non-dispersive limit,  $\delta$ -functions and their derivatives must be permitted so that  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  may become a (quasi-)local integral kernel. In the remainder of the paper, we will use the superscripts  $\top$  and  $+$  to indicate transposition and Hermitian conjugation with respect to tensor indices. Since the spatial arguments are not switched by these operations, the operator associated with an integral kernel  $\vec{A}(\mathbf{r}, \mathbf{r}')$  is Hermitian if  $\vec{A}(\mathbf{r}, \mathbf{r}') = \vec{A}^+(\mathbf{r}', \mathbf{r})$ . In particular, an operator associated with a real kernel is Hermitian if it has the reciprocity property  $\vec{A}(\mathbf{r}, \mathbf{r}') = \vec{A}^\top(\mathbf{r}', \mathbf{r})$ . The decomposition  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) = \text{Re } \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) + i \text{Im } \vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  of the conductivity tensor is therefore identical with the decomposition of the associated operator into a Hermitian and an anti-Hermitian part,

$$\begin{aligned} \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) &\equiv \text{Re } \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \\ &= \frac{1}{2} [\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) + \vec{Q}^+(\mathbf{r}', \mathbf{r}, \omega)], \end{aligned} \quad (2)$$

$$\begin{aligned} \vec{\tau}(\mathbf{r}, \mathbf{r}', \omega) &\equiv \text{Im } \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \\ &= \frac{1}{2i} [\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) - \vec{Q}^+(\mathbf{r}', \mathbf{r}, \omega)]. \end{aligned} \quad (3)$$

Since  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is associated with the dissipation of electromagnetic energy (see, e.g., Refs. [7, 14]), the operator associated with the integral kernel  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is, for real

$\omega$ , a positive definite operator in the case of absorbing media considered throughout this paper.

The conductivity tensor  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is the temporal Fourier transform of a response function  $\vec{Q}(\mathbf{r}, \mathbf{r}', t)$  in the time domain,

$$\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) = \int dt e^{i\omega t} \vec{Q}(\mathbf{r}, \mathbf{r}', t), \quad (4)$$

which satisfies causality conditions of the type

$$\vec{Q}(\mathbf{r}, \mathbf{r}', t) = 0 \quad \text{if } t - \cos \eta |\mathbf{r} - \mathbf{r}'|/c < 0 \quad (5)$$

for chosen  $\mathbf{r}$  and  $\mathbf{r}'$  and arbitrary directional cosines  $\cos \eta$  ( $0 \leq \cos \eta \leq 1$ ). In particular, for  $\cos \eta = 0$ , one finds from arguments [14–16] similar to those for the case of spatially locally responding media that, for chosen  $\mathbf{r}$  and  $\mathbf{r}'$ ,  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is analytic in the upper complex  $\omega$  half-plane, fulfills Kramers–Kronig (Hilbert transform) relations, and satisfies the Schwarz reflection principle  $\vec{Q}^*(\mathbf{r}, \mathbf{r}', \omega) = \vec{Q}(\mathbf{r}, \mathbf{r}', -\omega^*)$ . Other values of  $\cos \eta$  could obviously provide more stringent (spatio-temporal) conditions (see also Ref. [17]), which are, however, not required here.

Let us identify the current density that enters the macroscopic Maxwell equations in the frequency domain with  $\underline{\mathbf{j}}(\mathbf{r}, \omega)$  as specified in Eq. (1). In this case, the medium-assisted electric field in the frequency domain satisfies the integro-differential equation

$$\begin{aligned} \nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \underline{\mathbf{E}}(\mathbf{r}, \omega) \\ - i\mu_0\omega \int d^3r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{E}}(\mathbf{r}', \omega) = i\mu_0\omega \underline{\mathbf{j}}_N(\mathbf{r}, \omega), \end{aligned} \quad (6)$$

whose unique solution is

$$\underline{\mathbf{E}}(\mathbf{r}, \omega) = i\mu_0\omega \int d^3r' \vec{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{j}}_N(\mathbf{r}', \omega), \quad (7)$$

with  $\vec{G}(\mathbf{r}, \mathbf{r}', \omega)$  being the (retarded) Green tensor. It satisfies Eq. (6) with the (tensorial)  $\delta$ -function source,

$$\begin{aligned} \nabla \times \nabla \times \vec{G}(\mathbf{r}, \mathbf{s}, \omega) - \frac{\omega^2}{c^2} \vec{G}(\mathbf{r}, \mathbf{s}, \omega) \\ - i\mu_0\omega \int d^3r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \vec{G}(\mathbf{r}', \mathbf{s}, \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{s}), \end{aligned} \quad (8)$$

together with the boundary condition at infinity, and has all the attributes of a (Fourier transformed) causal response function just as  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  has them. In particular, it is analytic in the upper  $\omega$  half-plane and the Schwarz reflection principle  $\vec{G}^*(\mathbf{r}, \mathbf{r}', \omega) = \vec{G}(\mathbf{r}, \mathbf{r}', -\omega^*)$  is valid. Its basic properties in position space are similar to the ones known from the spatially local theory, in particular, since  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is reciprocal, so is  $\vec{G}(\mathbf{r}, \mathbf{r}', \omega)$ ,

$\vec{G}(\mathbf{r}, \mathbf{r}', \omega) = \vec{G}^\top(\mathbf{r}', \mathbf{r}, \omega)$ , and, for real  $\omega$ , the generalized integral relation

$$\begin{aligned} \mu_0 \omega \int d^3 s \int d^3 s' \vec{G}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{\sigma}(\mathbf{s}, \mathbf{s}', \omega) \cdot \vec{G}^*(\mathbf{s}', \mathbf{r}', \omega) \\ = \text{Im} \vec{G}(\mathbf{r}, \mathbf{r}', \omega) \end{aligned} \quad (9)$$

holds (App. A).

To quantize the theory, the Langevin noise source  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  is regarded as an operator  $[\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) \mapsto \hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)]$  with the commutation relation

$$[\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega), \hat{\mathbf{j}}_{\mathbf{N}}^\dagger(\mathbf{r}', \omega')] = \frac{\hbar \omega}{\pi} \delta(\omega - \omega') \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega). \quad (10)$$

The (now operator-valued) equation (7) relates the electric field operator

$$\hat{\mathbf{E}}(\mathbf{r}) = \int_0^\infty d\omega \hat{\mathbf{E}}(\mathbf{r}, \omega) + \text{H.c.}, \quad (11)$$

and thus all the electromagnetic field operators, to  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{j}}_{\mathbf{N}}^\dagger(\mathbf{r}', \omega')$ , which may be regarded as the dynamical variables of the overall system consisting of the electromagnetic field and the linear medium (incorporating the reservoir degrees of freedom responsible for absorption). It should be mentioned that, by means of the correspondence

$$\frac{i}{\varepsilon_0 \omega} \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \leftrightarrow \vec{\chi}(\mathbf{r}, \mathbf{r}', \omega), \quad (12)$$

where  $\vec{\chi}(\mathbf{r}, \mathbf{r}', \omega)$  is the (nonlocal) dielectric susceptibility tensor, the basic commutation relation (10) becomes equivalent to the commutation relation derived from a microscopic, linear two-band model of dielectric material [18], which has been used to study the quantized electromagnetic field in spatially dispersive dielectrics [8, 9].

In order to complete the quantization scheme, a Hamiltonian  $\hat{H}$  needs to be introduced [as a functional of  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{j}}_{\mathbf{N}}^\dagger(\mathbf{r}', \omega')$ ] so as to generate ‘free’ time evolution according to

$$[\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega), \hat{H}] = \hbar \omega \hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega), \quad (13)$$

which constrains the Hamiltonian to the form

$$\hat{H} = \pi \int_0^\infty d\omega \int d^3 r \int d^3 r' \hat{\mathbf{j}}_{\mathbf{N}}^\dagger(\mathbf{r}, \omega) \cdot \vec{\rho}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}', \omega), \quad (14)$$

to within irrelevant c-number contributions. A glance at Eqs. (10) and (13) now shows that  $\vec{\rho}(\mathbf{r}, \mathbf{r}', \omega)$  is the integral kernel of the inverse operator of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  (which exists). The validity of the quantization scheme is confirmed by checking that the well-known equal-time commutation relations for the electromagnetic field operators hold, which can be done, in analogy to the spatially local theory (cf. Refs. [19–21]), by properly taking into account the properties of  $\vec{G}(\mathbf{r}, \mathbf{r}', \omega)$  and  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  [in particular, Eq. (9)].

The Hamiltonian (14) may clearly be brought to the diagonal form

$$\hat{H} = \int d^3 r \int_0^\infty d\omega \hbar \omega \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega) \quad (15)$$

known from the spatially local theory, where  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  is a bosonic field,

$$[\hat{\mathbf{f}}(\mathbf{r}, \omega), \hat{\mathbf{f}}^\dagger(\mathbf{r}', \omega')] = \delta(\omega - \omega') \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \quad (16)$$

by performing a linear transformation of the variables, which we shall assume to be invertible. Writing

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \left( \frac{\hbar \omega}{\pi} \right)^{\frac{1}{2}} \int d^3 r' \vec{K}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}', \omega), \quad (17)$$

the diagonalization is achieved and Eqs. (16) and (10) are rendered equivalent if we choose the integral kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  such that, for real  $\omega$ , the integral equation

$$\int d^3 s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{K}^+(\mathbf{r}', \mathbf{s}, \omega) = \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) \quad (18)$$

holds, which is guaranteed to possess solutions (see Sec. III) since  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is the integral kernel of a positive definite operator.

So far we have considered the ‘free’ medium-assisted electromagnetic field. Its interaction with additional (e.g., atomic) systems can be included in the theory on the basis of the well-known minimal or multi-polar coupling schemes in the usual way (see, e.g., Ref. [22]).

### III. NATURAL VARIABLES AND PROJECTIVE VARIABLES

Let us now turn to the problem of constructing the integral kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. (18). For this purpose, we consider the eigenvalue problem

$$\int d^3 r' \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{F}(\alpha, \mathbf{r}', \omega) = \sigma(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \quad (19)$$

which, under appropriate regularity assumptions on the conductivity tensor  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$ , such as those listed below Eqs. (1), is well-defined. In particular, it features a real (positive) spectrum and a complete set of orthogonal eigensolutions, which we may take to be ( $\delta$ -)normalized. Note that the real  $\omega$  plays the role of a parameter here, and  $\alpha$  stands for the collection of (discrete and/or continuous) indices needed to label the eigenfunctions. Adopting a continuum notation, we may write

$$\int d\alpha \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \quad (20)$$

$$\int d^3 r \mathbf{F}^*(\alpha, \mathbf{r}, \omega) \cdot \mathbf{F}(\alpha', \mathbf{r}, \omega) = \delta(\alpha - \alpha'), \quad (21)$$

and the diagonal expansion of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  reads

$$\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \int d\alpha \sigma(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega), \quad (22)$$

which resembles the expansion of the dielectric susceptibility in the microscopic theory [18] mentioned above. Substituting Eq. (22) into Eq. (18), we may construct an integral kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  in the form of

$$\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \int d\alpha \sigma^{\frac{1}{2}}(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega), \quad (23)$$

where we choose  $\sigma^{1/2}(\alpha, \omega) > 0$  so that the operator associated with  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  is the positive, Hermitian square-root of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ . Obviously, this solution to Eq. (18) is not unique, since any other kernel of the form

$$\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) = \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}(\mathbf{s}, \mathbf{r}', \omega) \quad (24)$$

with  $\vec{V}(\mathbf{r}, \mathbf{s}, \omega)$  satisfying

$$\int d^3s \vec{V}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}^+(\mathbf{r}', \mathbf{s}, \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}') \quad (25)$$

also obeys Eq. (18). As we are interested in invertible transformations (17), the operator corresponding to  $\vec{V}(\mathbf{r}, \mathbf{s}, \omega)$  should be invertible as well, so that we can replace Eq. (25) with the stronger unitarity condition

$$\begin{aligned} & \int d^3s \vec{V}^+(\mathbf{s}, \mathbf{r}, \omega) \cdot \vec{V}(\mathbf{s}, \mathbf{r}', \omega) \\ &= \int d^3s \vec{V}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}^+(\mathbf{r}', \mathbf{s}, \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'). \end{aligned} \quad (26)$$

Without loss of generality (see App. B), we can base our further calculations on Eq. (23).

Inserting Eq. (23) into Eq. (17), we find that

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \left( \frac{\hbar\omega}{\pi} \right)^{\frac{1}{2}} \int d\alpha \sigma^{\frac{1}{2}}(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \hat{g}(\alpha, \omega), \quad (27)$$

where we have introduced the new variables

$$\hat{g}(\alpha, \omega) = \int d^3r \mathbf{F}^*(\alpha, \mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega), \quad (28)$$

referred to as the natural variables in the following. Needless to say that they are again of bosonic type,

$$[\hat{g}(\alpha, \omega), \hat{g}^\dagger(\alpha', \omega')] = \delta(\alpha - \alpha') \delta(\omega - \omega'). \quad (29)$$

Since the transformation (28) does not mix different  $\omega$  components, the Hamiltonian (15) is still diagonal when expressed in terms of the natural variables,

$$\hat{H} = \int d\alpha \int_0^\infty d\omega \hbar\omega \hat{g}^\dagger(\alpha, \omega) \hat{g}(\alpha, \omega), \quad (30)$$

as can be easily seen by inverting Eq. (28),

$$\hat{\mathbf{f}}(\mathbf{r}, \omega) = \int d\alpha \mathbf{F}(\alpha, \mathbf{r}, \omega) \hat{g}(\alpha, \omega), \quad (31)$$

and combining with Eq. (15), on recalling Eq. (21).

Let us organize the set of eigenfunctions  $\mathbf{F}(\alpha, \mathbf{r}, \omega)$  into (a discrete number of) subsets labeled by  $\lambda$  ( $\lambda = 1, 2, \dots, \Lambda$ ). With the notation  $\alpha \mapsto (\lambda, \beta)$ , Eq. (31) then reads

$$\hat{\mathbf{f}}(\mathbf{r}, \omega) = \sum_{\lambda} \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \quad (32)$$

where

$$\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega) = \int d\beta \mathbf{F}_{\lambda}(\beta, \mathbf{r}, \omega) \hat{g}_{\lambda}(\beta, \omega). \quad (33)$$

The operators associated with the integral kernels

$$\vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) = \int d\beta \mathbf{F}_{\lambda}(\beta, \mathbf{r}, \omega) \mathbf{F}_{\lambda}^*(\beta, \mathbf{r}', \omega) \quad (34)$$

form a complete set of orthogonal projectors. Obviously, these projectors and the operators associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  and  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  as given by Eq. (23) are commuting quantities. It is not difficult to see that the variables

$$\begin{aligned} \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega) &= \int d^3r' \vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}', \omega) \\ &= \int d\beta \mathbf{F}_{\lambda}(\beta, \mathbf{r}, \omega) \hat{g}_{\lambda}(\beta, \omega), \end{aligned} \quad (35)$$

referred to as projective variables in the following, obey the non-bosonic commutation relation

$$[\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \hat{\mathbf{f}}_{\lambda'}^\dagger(\mathbf{r}', \omega')] = \delta_{\lambda\lambda'} \delta(\omega - \omega') \vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega), \quad (36)$$

and the Hamiltonian (15) expressed in terms of the projective variables reads as

$$\hat{H} = \sum_{\lambda} \int d^3r \int_0^\infty d\omega \hbar\omega \hat{\mathbf{f}}_{\lambda}^\dagger(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega). \quad (37)$$

From Eqs.(36) and (37) it then follows that

$$\begin{aligned} [\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \hat{H}] &= \hbar\omega \int d^3r' \vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}', \omega) \\ &= \hbar\omega \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega). \end{aligned} \quad (38)$$

Inserting Eq. (32) in Eq. (17), we obtain

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \sum_{\lambda} \hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega), \quad (39)$$

where the  $\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega)$  are given by

$$\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega) = \left( \frac{\hbar\omega}{\pi} \right)^{\frac{1}{2}} \int d^3r' \vec{K}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}', \omega), \quad (40)$$

with

$$\begin{aligned}\vec{K}_\lambda(\mathbf{r}, \mathbf{r}', \omega) &= \int d^3s \vec{P}_\lambda(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{K}(\mathbf{s}, \mathbf{r}', \omega) \\ &= \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{P}_\lambda(\mathbf{s}, \mathbf{r}', \omega).\end{aligned}\quad (41)$$

Recalling Eq. (36), we can easily see that

$$[\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega), \hat{\mathbf{j}}_{\mathbf{N}\lambda'}^\dagger(\mathbf{r}', \omega')] = \frac{\hbar\omega}{\pi} \delta_{\lambda\lambda'} \delta(\omega - \omega') \vec{\sigma}_\lambda(\mathbf{r}, \mathbf{r}', \omega), \quad (42)$$

where  $\vec{\sigma}_\lambda(\mathbf{r}, \mathbf{r}', \omega)$  is defined according to Eq. (41) with  $\vec{\sigma}$  in place of  $\vec{K}$ . Summation of Eq. (42) over  $\lambda$  and  $\lambda'$  leads back to Eq. (10), so that the two equations are equivalent.

At this stage, we observe that there is the option to base the quantization scheme directly on Eqs. (37), (39), and (40), regarding the variables  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r}, \omega)$  as the basic dynamical variables of the theory and assigning to them bosonic commutation relations

$$[\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega), \hat{\mathbf{f}}_{\lambda'}^\dagger(\mathbf{r}', \omega')] = \delta_{\lambda\lambda'} \delta(\omega - \omega') \vec{I} \delta(\mathbf{r} - \mathbf{r}') \quad (43)$$

in place of Eq. (36). Note that, in so doing, back reference from the variables  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  to the original variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  is not possible anymore. As can be seen from Eqs. (40) and (41), Eq. (42) is satisfied also when the  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r}, \omega)$  are considered as bosonic variables, from which it follows [via Eq. (39)] that Eq. (10) also still holds and, as before, this implies that the correct electromagnetic-field commutation relations hold. The second line of Eq. (38) remains of course also true so that the correct time evolution is ensured as well.

Since the state space attributed to the bosonic variables  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r}, \omega)$  is, in general, different from the state space attributed to the original variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$  [or, equivalently, attributed to  $\hat{g}_\lambda(\beta, \omega)$  and  $\hat{g}_\lambda^\dagger(\beta, \omega)$ ], the allowable states must be restricted, by ruling out certain coherent superpositions of states in the sense of a super-selection rule. In App. C, we show that the condition imposed on the states may be described by means of a set of projectors  $\hat{P}_\lambda$  such that the allowable states  $|\psi\rangle$  can be characterized by

$$\hat{P}_\lambda |\psi\rangle = |\psi\rangle \quad \forall \lambda, \quad (44)$$

where the action of the projectors  $\hat{P}_\lambda$  in state space is closely related to the action of the projectors associated with the kernels (34) in position space. As a result, if the total Hamiltonian  $\hat{H}_{\text{tot}}$  composed of the Hamiltonian (37) and possible interaction terms (in the case where additional, active sources are present) commutes with all of the projectors  $\hat{P}_\lambda$ ,

$$[\hat{P}_\lambda, \hat{H}_{\text{tot}}] = 0 \quad \forall \lambda, \quad (45)$$

then allowable states remain allowable in the course of time, and the option of treating the  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r}, \omega)$

as bosonic variables can be safely exercised. Clearly, all the observables of interest should then also commute with the  $\hat{P}_\lambda$  so that no transition matrix elements between states belonging to different subspaces, i.e., between spaces attributed to different  $\lambda$  values, can ever come into play.

One can also consider decompositions of  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$ , where in place of the  $\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega)$  introduced above other quantities  $\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega)$  subject to the condition

$$\sum_\lambda \hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega) = \sum_\lambda \hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega) \quad (46)$$

are introduced, whose commutation relations may be quite different from those of the  $\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega)$ . Obviously, the total noise current density  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  as defined by Eq. (39) and the commutation relation (10) are not changed by such a transformation, briefly referred to as gauge transformation in the following. Moreover, since, with regard to Eq. (10), only the sum of the commutators  $[\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega), \hat{\mathbf{j}}_{\mathbf{N}\lambda'}^\dagger(\mathbf{r}', \omega')]$  over all  $\lambda$  and  $\lambda'$  is relevant, every chosen set of (algebraically consistent) commutators  $[\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega), \hat{\mathbf{j}}_{\mathbf{N}\lambda'}^\dagger(\mathbf{r}', \omega')]$  which leads to Eq. (10) yields, in principle, a consistent quantization scheme in its own right. A ‘substructure below’ Eq. (10) can hence be introduced with some arbitrariness, but since the various available alternatives are not necessarily equivalent to each other, a specific one should not be favored in the absence of good (physical) motivation. In contrast, if the observables of interest—including the Hamiltonian—can be viewed as functionals of  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  [rather than of the individual  $\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega)$ ], Eqs. (10) and (14) can be regarded, in view of the fluctuation-dissipation theorem(s) (see, e.g., Ref. [14]), as being unique, and hence, as invariable fundament of the theory.

From the above, it may be reasonable to widen the notion of projective variables as follows. If, for a chosen (physically motivated) decomposition of the noise current density, it is possible to (linearly) relate the  $\hat{\mathbf{j}}_{\mathbf{N}\lambda}(\mathbf{r}, \omega)$  in Eq. (46) to (new) variables  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  such that, upon considering the latter as bosonic variables, the validity of the basic equations (10) and (13) is ensured, then the specific quantization scheme so obtained may be regarded as arising from the general quantization scheme by excluding certain types of (superposition) states from state space, and restricting the dynamics (as well as the allowable observables) accordingly. The  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  may then be seen as projective variables in a wider sense.

#### IV. DIFFERENT CLASSES OF MEDIA

We proceed to show that rather different classes of media (usually studied separately) fit into the general quantization scheme developed in Sec. II. The main task to be performed is solving the eigenvalue problem (19), which

requires knowledge of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  for the specific medium under consideration. In two limiting cases, the exact solution to Eq. (19) can be given straightforwardly, namely, in the case of an inhomogeneous medium without spatial dispersion and in the case of a homogeneous medium that shows spatial dispersion. Let us, therefore, first examine these two cases in detail before considering more general situations.

### A. Spatially non-dispersive inhomogeneous media

The complete neglect of spatial dispersion means to regard the medium response, i.e.,  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$ , as being strictly local. If this is assumed, we have

$$\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \vec{\sigma}(\mathbf{r}, \omega) \delta(\mathbf{r} - \mathbf{r}'), \quad (47)$$

where  $\vec{\sigma}(\mathbf{r}, \omega)$  can be written in diagonal form as

$$\vec{\sigma}(\mathbf{r}, \omega) = \sum_{i=1}^3 \sigma_i(\mathbf{r}, \omega) \mathbf{e}_i(\mathbf{r}, \omega) \mathbf{e}_i^*(\mathbf{r}, \omega), \quad (48)$$

with  $\mathbf{e}_i(\mathbf{r}, \omega)$  ( $i = 1, 2, 3$ ) being orthonormal unit vectors. Hence, the eigenvalues  $\sigma(\alpha, \omega)$  and eigenfunctions  $\mathbf{F}(\alpha, \mathbf{r}, \omega)$  of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  read  $[\alpha \mapsto (i, \mathbf{s})] \sigma_i(\mathbf{s}, \omega)$  and

$$\mathbf{F}_i(\mathbf{s}, \mathbf{r}, \omega) = \mathbf{e}_i(\mathbf{s}, \omega) \delta(\mathbf{s} - \mathbf{r}), \quad (49)$$

respectively. Equation (23) then becomes

$$\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \vec{K}(\mathbf{r}, \omega) \delta(\mathbf{r} - \mathbf{r}'), \quad (50)$$

where

$$\vec{K}(\mathbf{r}, \omega) = \sum_{i=1}^3 \sigma_i^{1/2}(\mathbf{r}, \omega) \mathbf{e}_i(\mathbf{r}, \omega) \mathbf{e}_i^*(\mathbf{r}, \omega), \quad (51)$$

and Eq. (17) takes the form

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \left( \frac{\hbar \omega}{\pi} \right)^{\frac{1}{2}} \vec{K}(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega), \quad (52)$$

which just yields the well-known quantization scheme for a locally responding, possibly anisotropic dielectric material [19, 20], upon identifying  $\vec{\sigma}(\mathbf{r}, \omega) = \varepsilon_0 \omega \text{Im} \vec{\chi}(\mathbf{r}, \omega)$ , with  $\vec{\chi}(\mathbf{r}, \omega)$  being the (local) dielectric susceptibility tensor [cf. Eq. (12)]. The natural variables  $\hat{g}_i(\mathbf{r}, \omega)$  are here simply the components of  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  along the principal axes of the medium, which may in general vary with position and frequency,

$$\hat{g}_i(\mathbf{r}, \omega) = \mathbf{e}_i^*(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega), \quad (53)$$

$$\hat{\mathbf{f}}(\mathbf{r}, \omega) = \sum_{i=1}^3 \mathbf{e}_i(\mathbf{r}, \omega) \hat{g}_i(\mathbf{r}, \omega). \quad (54)$$

Identifying the index  $\lambda$  introduced in Eq. (32) with  $i$  and assuming that  $\sigma_i(\mathbf{r}, \omega) \neq \sigma_{i'}(\mathbf{r}, \omega)$  for  $i \neq i'$ , one can define, according to Eq. (34), the three projection kernels

$$\vec{P}_i(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{e}_i(\mathbf{r}, \omega) \mathbf{e}_i^*(\mathbf{r}', \omega) \delta(\mathbf{r} - \mathbf{r}'), \quad (55)$$

which, according to Eq. (35), give rise to three sets of projective variables,

$$\hat{\mathbf{f}}_i(\mathbf{r}, \omega) = \mathbf{e}_i(\mathbf{r}, \omega) \hat{g}_i(\mathbf{r}, \omega). \quad (56)$$

As long as the projective variables are not coupled to each other—which is obviously the case for the ‘free’ system governed by the Hamiltonian (38)—they can be regarded as being of bosonic type. In this case, instead of using the original set of bosonic variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$ , one can use three sets of bosonic variables  $\hat{\mathbf{f}}_i(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_i^\dagger(\mathbf{r}, \omega)$  associated with the three principal axes of the dielectric medium at each space point.

If two of the three eigenvalues  $\sigma_i(\mathbf{r}, \omega)$  coincide (uniaxial medium), the two corresponding projection kernels  $\vec{P}_i(\mathbf{r}, \mathbf{r}', \omega)$  should be combined into one projector (projecting on the plane perpendicular to the distinguished axis of the medium), thereby reducing the number of sets of projective variables to two. Clearly, if the three eigenvalues  $\sigma_i(\mathbf{r}, \omega)$  all coincide (isotropic medium), the three projection kernels  $\vec{P}_i(\mathbf{r}, \mathbf{r}', \omega)$  should be combined to give the unit kernel  $\vec{I} \delta(\mathbf{r} - \mathbf{r}')$ , corresponding to the use of the original variables.

### B. Spatially dispersive homogeneous media

In the limiting case of an (infinitely extended) homogeneous medium,  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is translationally invariant, i.e., it is a function of the difference  $\mathbf{r} - \mathbf{r}'$ , and so is then  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ . We may therefore represent it as the spatial Fourier transform

$$\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{\sigma}(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}, \quad (57)$$

where

$$\vec{\sigma}(\mathbf{k}, \omega) = \sum_{i=1}^3 \sigma_i(\mathbf{k}, \omega) \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega), \quad (58)$$

with  $\mathbf{e}_i(\mathbf{k}, \omega)$  ( $i = 1, 2, 3$ ) being orthogonal unit vectors. Consequently, the eigenvalues  $\sigma(\alpha, \omega)$  and eigenfunctions  $\mathbf{F}(\alpha, \mathbf{r}, \omega)$  of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  are  $[\alpha \mapsto (i, \mathbf{k})] \sigma_i(\mathbf{k}, \omega)$  and

$$\mathbf{F}_i(\mathbf{k}, \mathbf{r}, \omega) = (2\pi)^{-3/2} e^{i\mathbf{k} \cdot \mathbf{r}} \mathbf{e}_i(\mathbf{k}, \omega), \quad (59)$$

respectively, and Eq. (23) reads

$$\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{K}(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}, \quad (60)$$

where

$$\vec{K}(\mathbf{k}, \omega) = \sum_{i=1}^3 \sigma_i^{1/2}(\mathbf{k}, \omega) \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega). \quad (61)$$

Combination of Eqs. (17), (60), and (61) then yields

$$\begin{aligned} \mathbf{j}_N(\mathbf{r}, \omega) &= \left( \frac{\hbar \omega}{\pi} \right)^{\frac{1}{2}} \frac{1}{(2\pi)^{3/2}} \\ &\times \sum_{i=1}^3 \int d^3k e^{i\mathbf{k}\cdot\mathbf{r}} \sigma_i^{1/2}(\mathbf{k}, \omega) \mathbf{e}_i(\mathbf{k}, \omega) \hat{g}_i(\mathbf{k}, \omega), \end{aligned} \quad (62)$$

where the natural variables  $\hat{g}_i(\mathbf{k}, \omega)$  are related to the spatial Fourier components of  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  as

$$\hat{g}_i(\mathbf{k}, \omega) = \frac{1}{(2\pi)^{3/2}} \int d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} \mathbf{e}_i^*(\mathbf{k}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega). \quad (63)$$

On the basis of the three unit vectors  $\mathbf{e}_i(\mathbf{k}, \omega)$ , three (different) projection kernels can be introduced,

$$\vec{P}_i(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \quad (64)$$

provided that  $\sigma_i(\mathbf{k}, \omega) \neq \sigma_{i'}(\mathbf{k}, \omega)$  for  $i \neq i'$ .

Let us consider, in particular, isotropic media that in addition do not feature optical activity in more detail. In this case, the diagonal form of the tensor  $\vec{\sigma}(\mathbf{k}, \omega)$  reads (see Ref. [7])

$$\vec{\sigma}(\mathbf{k}, \omega) = \sigma_{\parallel}(k, \omega) \frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\perp}(k, \omega) \left( \vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right), \quad (65)$$

i.e.,  $\sigma_1(k, \omega) = \sigma_{\parallel}(k, \omega)$  and  $\sigma_2(k, \omega) = \sigma_3(k, \omega) = \sigma_{\perp}(k, \omega) \neq \sigma_{\parallel}(k, \omega)$ , which implies that  $\vec{K}(\mathbf{k}, \omega)$ , Eq. (61), takes the form

$$\vec{K}(\mathbf{k}, \omega) = \sigma_{\parallel}^{1/2}(k, \omega) \frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\perp}^{1/2}(k, \omega) \left( \vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right). \quad (66)$$

Thus, the well-known longitudinal and transverse tensorial  $\delta$ -functions  $\vec{\Delta}_{\parallel}(\mathbf{r} - \mathbf{r}')$  and  $\vec{\Delta}_{\perp}(\mathbf{r} - \mathbf{r}')$ , respectively, can be taken as projection kernels,

$$\vec{P}_{\parallel(\perp)}(\mathbf{r}, \mathbf{r}', \omega) = \vec{\Delta}_{\parallel(\perp)}(\mathbf{r} - \mathbf{r}'), \quad (67)$$

which may be used to introduce, according to Eq. (35), the projective variables

$$\hat{\mathbf{f}}_{\parallel(\perp)}(\mathbf{r}, \omega) = \int d^3s \vec{\Delta}_{\parallel(\perp)}(\mathbf{r} - \mathbf{s}) \cdot \hat{\mathbf{f}}(\mathbf{s}, \omega). \quad (68)$$

### 1. Unitarily equivalent formulation

As already pointed out in Sec. II, the integral kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. (17) is not uniquely determined by

Eq. (18), since any other kernel  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  of the form (24) [together with Eq. (26)] is also an allowed kernel. To illustrate this for the isotropic medium under study, we first note that Eq. (65) may be equivalently rewritten as

$$\vec{\sigma}(\mathbf{k}, \omega) = \sigma_{\parallel}(k, \omega) \vec{I} - \mathbf{k} \times \gamma(k, \omega) \vec{I} \times \mathbf{k}, \quad (69)$$

where

$$\gamma(k, \omega) = [\sigma_{\perp}(k, \omega) - \sigma_{\parallel}(k, \omega)]/k^2. \quad (70)$$

Since (for real  $\omega$ )  $\sigma_{\parallel}(k, \omega)$  and  $\sigma_{\perp}(k, \omega)$  are both real and positive [in accordance with the requirement that  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  be the integral kernel of a positive definite operator],  $\gamma(k, \omega)$  is real but its sign is not determined by this requirement. However, if  $\gamma(k, \omega)$  is required here and below to be positive throughout, then

$$\vec{K}'(\mathbf{k}, \omega) = \sigma_{\parallel}^{1/2}(k, \omega) \vec{I} \pm \gamma^{1/2}(k, \omega) \mathbf{k} \times \vec{I} \quad (71)$$

obeys the equation

$$\vec{K}'(\mathbf{k}, \omega) \cdot \vec{K}'^+(\mathbf{k}, \omega) = \vec{\sigma}(\mathbf{k}, \omega). \quad (72)$$

Moreover, it can be shown that  $\vec{K}'(\mathbf{k}, \omega)$  can be represented in the form

$$\vec{K}'(\mathbf{k}, \omega) = \vec{K}(\mathbf{k}, \omega) \cdot \vec{V}(\mathbf{k}, \omega), \quad (73)$$

with

$$\begin{aligned} \vec{V}(\mathbf{k}, \omega) &= \frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\parallel}^{1/2}(k, \omega) \sigma_{\perp}^{-1/2}(k, \omega) \left( \vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right) \\ &\pm \gamma^{1/2}(k, \omega) \sigma_{\perp}^{-1/2}(k, \omega) \mathbf{k} \times \vec{I} \end{aligned} \quad (74)$$

$[\vec{V}^{-1}(\mathbf{k}, \omega) = \vec{V}^+(\mathbf{k}, \omega)]$ . Hence,  $\vec{K}'(\mathbf{k}, \omega)$  also yields, according to Eq. (60), a valid integral kernel  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$ ,

$$\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{K}'(\mathbf{k}, \omega) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \quad (75)$$

which is related to the integral kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  according to Eq. (24), where

$$\vec{V}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{V}(\mathbf{k}, \omega) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \quad (76)$$

with the associated operator being unitary. We thus see that the two formulations of the theory based on  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  and  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$ , respectively, are unitarily equivalent. Note that  $\vec{K}'(\mathbf{k}, \omega) \neq \vec{K}'^+(\mathbf{k}, \omega)$ , so that the operator associated with the integral kernel  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  is non-Hermitian (as to be expected, see App. B). Since the operators associated with  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  [as well as  $\vec{V}(\mathbf{r}, \mathbf{r}', \omega)$ ] and  $\vec{P}_{\parallel(\perp)}(\mathbf{r}, \mathbf{r}', \omega)$  commute, the same projectors may be employed in the two formulations of the theory to introduce projective variables according to Eq. (68).

## 2. Local limit: magnetodielectric media

Now let us suppose that  $\sigma_{\parallel}(k, \omega)$  and  $\gamma(k, \omega)$  in Eq. (69) are sufficiently slowly varying functions of  $k$ , with well-defined and unique long-wavelength limits  $\lim_{k \rightarrow 0} \sigma_{\parallel}(k, \omega) = \sigma_{\parallel}(\omega) > 0$  and  $\lim_{k \rightarrow 0} \gamma(k, \omega) = \gamma(\omega) > 0$ , so that they may be approximated by these limits under the integral in Eq. (57) to obtain

$$\begin{aligned} \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad - \gamma(\omega) \nabla \times [\vec{I} \delta(\mathbf{r} - \mathbf{r}')] \times \vec{\nabla}'. \end{aligned} \quad (77)$$

It should be pointed out that in the limiting case given by Eq. (77) the positive definiteness of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  already implies that  $\gamma(\omega)$  must be positive,  $\gamma(\omega) > 0$ ; in the general case as given by Eq. (57) together with Eqs. (69) and (70), the positive definiteness of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  does not automatically restrict  $\gamma(k, \omega)$  to positive values.

In order to see to what type of medium this  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  corresponds, we have to find from Eq. (77) the full conductivity tensor  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$ , which is uniquely possible since Eqs. (2) and (3) are Hilbert transforms of each other (cf. Sec. II). The full conductivity tensor corresponding to Eq. (77) is thus of the form

$$\begin{aligned} \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) &= Q^{(1)}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad - Q^{(2)}(\omega) \nabla \times [\vec{I} \delta(\mathbf{r} - \mathbf{r}')] \times \vec{\nabla}', \end{aligned} \quad (78)$$

where  $Q^{(1)}(\omega)$  and  $Q^{(2)}(\omega)$  are (Fourier-transformed) response functions, both of which are determined by their respective real parts  $\sigma_{\parallel}(\omega)$  and  $\gamma(\omega)$ . Inserting Eq. (78) into Eq. (1) and comparing with

$$\begin{aligned} \hat{\mathbf{j}}(\mathbf{r}, \omega) &= -i\varepsilon_0\omega [\varepsilon(\omega) - 1] \hat{\mathbf{E}}(\mathbf{r}, \omega) \\ &\quad + \kappa_0 \nabla \times \{[1 - \kappa(\omega)] \hat{\mathbf{B}}(\mathbf{r}, \omega)\} + \hat{\mathbf{j}}_{\text{N}}(\mathbf{r}, \omega) \end{aligned} \quad (79)$$

$[\hat{\mathbf{B}}(\mathbf{r}, \omega) = (i\omega)^{-1} \nabla \times \hat{\mathbf{E}}(\mathbf{r}, \omega)]$ , which is the well-known description of a locally responding (homogeneous) magnetodielectric medium, we can make the identifications

$$Q^{(1)}(\omega) = -i\varepsilon_0\omega [\varepsilon(\omega) - 1] \quad (80)$$

and

$$Q^{(2)}(\omega) = -i\kappa_0[1 - \kappa(\omega)]/\omega, \quad (81)$$

where  $\varepsilon(\omega)$  is the permittivity and  $\mu(\omega) = \kappa^{-1}(\omega)$  the (paramagnetic) permeability of the medium ( $\mu_0 = \kappa_0^{-1}$ ). For real  $\omega$ , we thus obtain

$$\sigma_{\parallel}(\omega) = \varepsilon_0\omega \text{Im} \varepsilon(\omega) \quad (82)$$

and

$$\gamma(\omega) = -\kappa_0 \text{Im} \kappa(\omega)/\omega. \quad (83)$$

Note that, because of  $\gamma(\omega) > 0$ , from Eq. (83) it follows that  $\text{Im} \kappa(\omega) < 0$  for  $\omega > 0$ , from which it can be shown that  $\mu(\omega \rightarrow 0) > 1$ .

At first glance, one might believe (erroneously) that not only paramagnetic [ $\mu(\omega \rightarrow 0) > 1$ ] but also diamagnetic [ $\mu(\omega \rightarrow 0) < 1$ ] features of a medium (or the combined effect of both) can be consistently described by means of the magnetic permeability  $\mu(\omega)$  which is included, as seen above, in the basic linear-response constitutive relation (1). However, since diamagnetism is basically a nonlinear effect (as the underlying microscopic Hamiltonian is quadratic in the magnetic induction field), it is beyond the scope of linear response theory. If it is desired to include diamagnetic media in the framework of linear electrodynamics nevertheless, one can regard the magnetic field on which the diamagnetic susceptibility depends as being (the mean value of) an externally controlled field independent of the dynamical variables. Note that the Onsager reciprocity theorem needs to be stated in its generalized form in this case, see Refs. [7, 14]. For a more satisfactory account of diamagnetic media, one should, however, resort to a non-linear response formalism, or to a more microscopic theory.

An obvious solution to Eq. (18) with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  given by Eq. (77) is provided by

$$\begin{aligned} \vec{K}'(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}^{1/2}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \mp i\gamma^{1/2}(\omega) \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \end{aligned} \quad (84)$$

which corresponds to the kernel (75) [together with Eq. (71)] when, for an isotropic medium,  $\sigma_{\parallel}(k, \omega)$  and  $\gamma(k, \omega)$  are approximated by  $\sigma_{\parallel}(\omega)$  and  $\gamma(\omega)$ , respectively. The kernel (75) [together with Eq. (71)] fits well here since it depends in a particularly simple way on those quantities that we have assumed to approach well-defined limits in the derivation that led to Eq. (77), a property which can be attributed to the responsible transformation (76) [together with (74)]. In contrast, the kernel obtained directly from Eq. (60) [together with Eq. (66)], by first eliminating  $\sigma_{\perp}(k, \omega)$  by means of Eq. (70) and then approximating  $\sigma_{\parallel}(k, \omega) \mapsto \sigma_{\parallel}(\omega)$  and  $\gamma(k, \omega) \mapsto \gamma(\omega)$ , does not provide an alternative to Eq. (84), as it does not lead to Eq. (77) when inserted in Eq. (18); it corresponds to a different medium. Correspondingly, the kernel  $\vec{V}(\mathbf{r}, \mathbf{r}', \omega)$  obtained by expressing Eq. (76) [with Eq. (74)] in terms of  $\sigma_{\parallel}(k, \omega)$  and  $\gamma(k, \omega)$  and then approximating them by  $\sigma_{\parallel}(\omega)$  and  $\gamma(\omega)$ , respectively, fails to be unitary.

Substituting for  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. (17)  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  as given by Eq. (84) [together with Eqs. (82) and (83)], we may explicitly express the noise current density in terms of the bosonic dynamical variables to obtain

$$\begin{aligned} \hat{\mathbf{j}}_{\text{N}}(\mathbf{r}, \omega) &= \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{\frac{1}{2}} \sqrt{\omega^2 \text{Im} \varepsilon(\omega)} \hat{\mathbf{f}}(\mathbf{r}, \omega) \\ &\quad \mp i \left(\frac{\hbar\kappa_0}{\pi}\right)^{\frac{1}{2}} \nabla \times [\sqrt{-\text{Im} \kappa(\omega)} \hat{\mathbf{f}}(\mathbf{r}, \omega)]. \end{aligned} \quad (85)$$



Since the operators associated with the projection kernels (67) commute with the operators associated with Eqs. (84) and (77), one may introduce the projective variables  $\hat{\mathbf{f}}_{\parallel(\perp)}(\mathbf{r}, \omega)$  defined by Eq. (68), which corresponds to a decomposition of  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  into longitudinal and transverse parts,

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \hat{\mathbf{j}}_{\mathbf{N}\parallel}(\mathbf{r}, \omega) + \hat{\mathbf{j}}_{\mathbf{N}\perp}(\mathbf{r}, \omega), \quad (86)$$

where

$$\hat{\mathbf{j}}_{\mathbf{N}\parallel}(\mathbf{r}, \omega) = \left( \frac{\hbar \varepsilon_0}{\pi} \right)^{\frac{1}{2}} \sqrt{\omega^2 \text{Im} \varepsilon(\omega)} \hat{\mathbf{f}}_{\parallel}(\mathbf{r}, \omega), \quad (87)$$

$$\begin{aligned} \hat{\mathbf{j}}_{\mathbf{N}\perp}(\mathbf{r}, \omega) &= \left( \frac{\hbar \varepsilon_0}{\pi} \right)^{\frac{1}{2}} \sqrt{\omega^2 \text{Im} \varepsilon(\omega)} \hat{\mathbf{f}}_{\perp}(\mathbf{r}, \omega) \\ &\mp i \left( \frac{\hbar \kappa_0}{\pi} \right)^{\frac{1}{2}} \nabla \times [\sqrt{-\text{Im} \kappa(\omega)} \hat{\mathbf{f}}_{\perp}(\mathbf{r}, \omega)]. \end{aligned} \quad (88)$$

Making use of Eq. (35) and identifying therein the projection kernels  $\vec{P}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega)$  with  $\vec{\Delta}_{\parallel(\perp)}(\mathbf{r} - \mathbf{r}')$ , one may then proceed as described in Sec. III and regard the projective variables  $\hat{\mathbf{f}}_{\parallel}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_{\perp}(\mathbf{r}, \omega)$  as being two independent sets of bosonic variables.

Let us briefly make contact with the quantization scheme described in Refs. [19, 21], where  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$  is decomposed according to

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \hat{\mathbf{j}}_{\mathbf{N}e}(\mathbf{r}, \omega) + \hat{\mathbf{j}}_{\mathbf{N}m}(\mathbf{r}, \omega), \quad (89)$$

with

$$\hat{\mathbf{j}}_{\mathbf{N}e}(\mathbf{r}, \omega) = \left( \frac{\hbar \varepsilon_0}{\pi} \right)^{\frac{1}{2}} \sqrt{\omega^2 \text{Im} \varepsilon(\omega)} \hat{\mathbf{f}}_e(\mathbf{r}, \omega), \quad (90)$$

$$\hat{\mathbf{j}}_{\mathbf{N}m}(\mathbf{r}, \omega) = \mp i \left( \frac{\hbar \kappa_0}{\pi} \right)^{\frac{1}{2}} \nabla \times [\sqrt{-\text{Im} \kappa(\omega)} \hat{\mathbf{f}}_m(\mathbf{r}, \omega)]. \quad (91)$$

The connection between Eqs. (86)–(88) and Eqs. (89)–(91) is given by a gauge transformation (cf. Sec. III), which effectively redistributes the first term of Eq. (88). It is not difficult to prove that the total noise current as given by Eq. (89) satisfies the correct commutation relation (10) [with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  from Eq. (77) together with Eqs. (82) and (83)] if  $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$  are regarded as two independent sets of bosonic variables. Since  $\hat{\mathbf{j}}_{\mathbf{N}e}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{j}}_{\mathbf{N}m}(\mathbf{r}, \omega)$  can be linearly related to  $\hat{\mathbf{j}}_{\mathbf{N}\parallel}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{j}}_{\mathbf{N}\perp}(\mathbf{r}, \omega)$  and thus to  $\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega)$ , the variables  $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$  may be viewed as projective variables, in the sense outlined at the end of Sec. III. Since Eq. (89) [with Eqs. (90) and (91)] is a separation of the noise current into a part attributed to a dielectric polarization and a part attributed to a (paramagnetic) magnetization, the quantization scheme based on Eqs. (89)–(91), with  $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$  being bosonic variables, may be thought of as following from the general quantization scheme in the case where magneto-electric crossing effects can be a priori excluded from consideration.

### 3. Local limit: other kinds of media

The transition to the local limit is not a unique procedure in general. Various kinds of locally responding (homogeneous) media, including non-isotropic ones, may therefore be derived as limiting cases from Eq. (57). To illustrate this, let us represent  $\vec{\sigma}(\mathbf{k}, \omega)$  as given in Eq. (58) in a different orthonormal basis, where the new expansion will be non-diagonal in general,

$$\vec{\sigma}(\mathbf{k}, \omega) = \sum_{i,j=1}^3 \tilde{\sigma}_{ij}(\mathbf{k}, \omega) \tilde{\mathbf{e}}_i(\mathbf{k}, \omega) \tilde{\mathbf{e}}_j^*(\mathbf{k}, \omega). \quad (92)$$

The new basis vectors  $\tilde{\mathbf{e}}_i(\mathbf{k}, \omega)$  are related to the ones appearing in Eq. (58) by a unitary transformation,

$$\tilde{\mathbf{e}}_i(\mathbf{k}, \omega) = \sum_{k=1}^3 U_{ik}(\mathbf{k}, \omega) \mathbf{e}_k(\mathbf{k}, \omega), \quad (93)$$

$$U_{ik}(\mathbf{k}, \omega) = \tilde{\mathbf{e}}_i(\mathbf{k}, \omega) \cdot \mathbf{e}_k^*(\mathbf{k}, \omega). \quad (94)$$

We may always choose the  $\tilde{\mathbf{e}}_i(\mathbf{k}, \omega)$  so that they are independent of  $\mathbf{k}$ ,  $\tilde{\mathbf{e}}_i(\mathbf{k}, \omega) \mapsto \tilde{\mathbf{e}}_i(\omega)$ . If this choice can be made such that the new expansion coefficients,

$$\tilde{\sigma}_{ij}(\mathbf{k}, \omega) = \sum_{k,l=1}^3 U_{ik}^*(\mathbf{k}, \omega) \sigma_{kl}(\mathbf{k}, \omega) U_{jl}(\mathbf{k}, \omega), \quad (95)$$

may be approximately replaced under the  $\mathbf{k}$ -integral according to

$$\tilde{\sigma}_{ij}(\mathbf{k}, \omega) \mapsto \tilde{\sigma}_{ij}(\mathbf{k} \rightarrow 0, \omega) \equiv \tilde{\sigma}_{ij}(\omega) \quad (96)$$

when Eq. (92) is inserted in Eq. (57), then in this way the type of locally responding (homogeneous) anisotropic medium defined by Eq. (47) is recovered. [Equation (48) is then obtained by diagonalizing  $\tilde{\sigma}_{ij}(\omega)$  by means of yet another ( $\mathbf{k}$ -independent) unitary transformation.] Similarly, if the approximation (96) is generalized to include further terms of an (assumed) expansion of  $\tilde{\sigma}_{ij}(\mathbf{k}, \omega)$  at  $\mathbf{k} = 0$ , then quasi-local approximations of Eq. (57) are generated, by inserting the truncated expansion into Eq. (57) and integrating term by term to yield a linear combination of various derivatives of  $\delta$ -functions. In pursuing such approximation procedures—whose validity is to be examined in each case and which depends crucially on the choice of the transformation (93), (94) (i.e., of the new basis vectors)—it must be kept in mind that any approximate form of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  so derived has to conform to all the general requirements on  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$ . In fact, the most general kind of linear medium discussed in terms of local constitutive relations in the literature, the so-called bi-anisotropic medium (see, e.g., Ref. [23]), may be viewed in this way as a quasi-local approximation of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  [and thereby of  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$ ] that incorporates derivatives of  $\delta$ -functions up to the second order.

### C. Spatially dispersive inhomogeneous media

As already mentioned, knowledge of the medium properties, i.e., of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ , is required in order to solve the eigenvalue problem (19) and perform explicitly the quantization of the medium-assisted electromagnetic field—a task which, in general, cannot be accomplished in closed form. Nevertheless, to provide some analytical insight into the problem, let us consider media that combine the features of the media considered in Secs. IV A and IV B in an approximate fashion.

#### 1. Model

We assume that the medium permits one to clearly distinguish between the length scales associated with spatial dispersion and inhomogeneity, with the former scale being sufficiently small as compared with the latter one. In this case, the medium can be regarded as having locally the properties of bulk material, and  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  may be approximated as

$$\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{\Omega} \sum_{\mathbf{L}} \sum_{i=1}^3 \sum_{\mathbf{k}} \sigma_{i\mathbf{L}\mathbf{k}}(\omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \times \theta_{\mathbf{L}}(\mathbf{r}) \theta_{\mathbf{L}}(\mathbf{r}') \mathbf{e}_{i\mathbf{L}\mathbf{k}}(\omega) \mathbf{e}_{i\mathbf{L}\mathbf{k}}^*(\omega), \quad (97)$$

from which the eigenfunctions of the associated operator are seen to be

$$\mathbf{F}_{i\mathbf{L}\mathbf{k}}(\mathbf{r}, \omega) = \Omega^{-1/2} \theta_{\mathbf{L}}(\mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{r}} \mathbf{e}_{i\mathbf{L}\mathbf{k}}(\omega). \quad (98)$$

Here, the medium is thought of as being divided into unit cells of volume  $\Omega$  which form a Bravais-type lattice, the cut-off function  $\theta_{\mathbf{L}}(\mathbf{r})$  is unity if  $\mathbf{r}$  is in the cell of lattice vector  $\mathbf{L}$  and zero otherwise,  $\mathbf{e}_{i\mathbf{L}\mathbf{k}}(\omega)$  are, for chosen  $\mathbf{L}$ ,  $\mathbf{k}$ , and  $\omega$ , a triplet ( $i = 1, 2, 3$ ) of orthogonal unit vectors, and the wave vector  $\mathbf{k}$  runs over the reciprocal lattice. Note that, for each cell  $\mathbf{L}$ ,

$$\vec{\sigma}_{\mathbf{L}\mathbf{k}}(\omega) = \sum_{i=1}^3 \sigma_{i\mathbf{L}\mathbf{k}}(\omega) \mathbf{e}_{i\mathbf{L}\mathbf{k}}(\omega) \mathbf{e}_{i\mathbf{L}\mathbf{k}}^*(\omega) \quad (99)$$

corresponds to the diagonal form in the  $(\mathbf{k}, \omega)$  domain of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  for bulk material [cf. Eq. (58)].

The main features of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  as given in Eq. (97) can be summarized as follows. (i)  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  is zero whenever  $\mathbf{r}$  and  $\mathbf{r}'$  are not in the same cell, so that  $\Omega^{1/3}$  determines the length scale on which spatial dispersion is at most observed. (ii) The dependence on  $\mathbf{L}$  of  $\vec{\sigma}_{\mathbf{L}\mathbf{k}}(\omega)$  [Eq. (99)] for an inhomogeneous medium introduces an  $\mathbf{L}$ -dependence into Eq. (97) which should be sufficiently weak, so that noticeable violations of the translational invariance of  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  may occur only on a length scale that is large compared with  $\Omega^{1/3}$ . Needless to say that the main features do not essentially change if  $\theta_{\mathbf{L}}(\mathbf{r})$  is

replaced by another—but qualitatively similar—cut-off function.

Let us denote by  $\mathbf{L}(\mathbf{r})$  the particular lattice vector whose cell contains the point  $\mathbf{r}$ , so that  $\mathbf{L}(\mathbf{r})$  plays the role of a coarse-grained position variable. With the notations  $\theta_{\mathbf{L}(\mathbf{r})}(\mathbf{r}') \mapsto \theta[\mathbf{L}(\mathbf{r}), \mathbf{r}']$  and  $\vec{\sigma}_{\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega) \mapsto \vec{\sigma}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}), \omega]$ , Eq. (97) together with Eq. (99) can be rewritten as

$$\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \theta[\mathbf{L}(\mathbf{r}'), \mathbf{r}] \vec{\sigma}[\mathbf{L}(\mathbf{r}'), \mathbf{r} - \mathbf{r}', \omega], \quad (100)$$

with

$$\vec{\sigma}[\mathbf{L}(\mathbf{r}'), \mathbf{r} - \mathbf{r}', \omega] = \frac{1}{\Omega} \sum_{\mathbf{k}} \vec{\sigma}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}'), \omega] e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}. \quad (101)$$

Note that for arbitrary (continuous) values  $\mathbf{s}$ , the function  $\theta(\mathbf{s}, \mathbf{r})$  can be regarded as being symmetric. Using Eq. (98), we find that Eq. (23) takes the form

$$\vec{K}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\theta[\mathbf{L}(\mathbf{r}'), \mathbf{r}]}{\Omega} \sum_{\mathbf{k}} \vec{K}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}'), \omega] e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}, \quad (102)$$

where

$$\vec{K}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}), \omega] = \sum_{i=1}^3 \sigma_{i\mathbf{k}}^{1/2}[\mathbf{L}(\mathbf{r}), \omega] \mathbf{e}_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}), \omega] \mathbf{e}_{i\mathbf{k}}^*[\mathbf{L}(\mathbf{r}), \omega] \quad (103)$$

$\{\sigma_{i\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega) \mapsto \sigma_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}), \omega], \mathbf{e}_{i\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega) \mapsto \mathbf{e}_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}), \omega]\}.$

It can be shown that Eq. (100) [with Eq. (101)] indeed contains (and, in a sense, interpolates) the two limiting cases studied in Secs. IV A and IV B. For the proof, we observe that in the case of negligible spatial dispersion, the cell size can be shrunk to zero,  $\Omega \rightarrow 0$ , so that the lattice vectors take on continuous values,  $\mathbf{L}(\mathbf{r}) \rightarrow \mathbf{r}$ . As the lattice becomes finer and finer, the reciprocal lattice becomes more and more coarse, and, for  $\mathbf{r}$  and  $\mathbf{r}'$  unequal but in the same cell, all the points of the reciprocal lattice with  $\mathbf{k} \neq 0$  give rise to rapidly oscillating terms in Eq. (101). In the limit  $\Omega \rightarrow 0$ , these terms oscillate infinitely rapidly and average to zero (when applying the operator associated with Eq. (100) [with Eq. (101)] to any reasonable function), so that they may be set equal to zero. Taking also into account that  $\theta[\mathbf{L}(\mathbf{r}'), \mathbf{r}]/\Omega \rightarrow \delta(\mathbf{r} - \mathbf{r}')$  in this limit, we see that Eq. (100) [with Eq. (101)] indeed approaches Eq. (47) for vanishing spatial dispersion [note the correspondences  $\sigma_{i\mathbf{k}=0}[\mathbf{L}(\mathbf{r}) = \mathbf{r}, \omega] = \sigma_i(\mathbf{r}, \omega)$  and  $\mathbf{e}_{i\mathbf{k}=0}[\mathbf{L}(\mathbf{r}) = \mathbf{r}, \omega] = \mathbf{e}_i(\mathbf{r}, \omega)$ ].

On the other hand, in the limiting case of an infinitely extended homogeneous medium, there is no  $\mathbf{L}$ -dependence of the medium properties so that we are free to increase the cell size indefinitely,  $\Omega \rightarrow \infty$ . Consequently, we may let  $\theta[\mathbf{L}(\mathbf{r}'), \mathbf{r}] \rightarrow 1$  in Eq. (100) and  $\vec{\sigma}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}), \omega] \rightarrow \vec{\sigma}(\mathbf{k}, \omega)$ ,  $\Omega^{-1} \sum_{\mathbf{k}} \rightarrow (2\pi)^{-3} \int d^3k$  in Eq. (101), which reveals that Eq. (100) [with Eq. (101)] approaches Eq. (57) as expected.

## 2. Magnetodielectric media

To quantize the electromagnetic field in an inhomogeneous magnetodielectric medium specified in terms of  $\varepsilon(\mathbf{r}, \omega)$  and  $\kappa(\mathbf{r}, \omega) = \mu^{-1}(\mathbf{r}, \omega)$ , let us consider a medium that is both sufficiently weakly inhomogeneous and sufficiently weakly spatially dispersive, so that  $\Omega$  in Eq. (101) can be chosen on a scale intermediate between the scales of spatial dispersion and inhomogeneity. We may then approximately let  $\mathbf{L}(\mathbf{r})$  be a continuous variable,  $\mathbf{L}(\mathbf{r}) \rightarrow \mathbf{r}$  in Eq. (100), and yet, at the same time, approximately treat the  $\mathbf{k}$ -sum in Eq. (101) as an integral, so that Eq. (100) [with Eq. (101)] approximates to  $[\theta(\mathbf{L}(\mathbf{r}'), \mathbf{r}) \rightarrow \theta(\mathbf{r}', \mathbf{r})]$

$$\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\theta(\mathbf{r}', \mathbf{r})}{(2\pi)^3} \int d^3k \vec{\sigma}(\mathbf{r}', \mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}. \quad (104)$$

For a medium that is locally of the type described by Eq. (69), we may set

$$\vec{\sigma}(\mathbf{r}, \mathbf{k}, \omega) = \sigma_{\parallel}(\mathbf{r}, k, \omega) \vec{I} - \mathbf{k} \times \gamma(\mathbf{r}, k, \omega) \vec{I} \times \mathbf{k}, \quad (105)$$

where

$$\gamma(\mathbf{r}, k, \omega) = [\sigma_{\perp}(\mathbf{r}, k, \omega) - \sigma_{\parallel}(\mathbf{r}, k, \omega)]/k^2 > 0. \quad (106)$$

Assuming that in the  $\mathbf{k}$ -integral in Eq. (104),  $\sigma_{\parallel}(\mathbf{r}, k, \omega)$  and  $\gamma(\mathbf{r}, k, \omega)$  may be approximated, respectively, by well-defined (and unique) long-wavelength limits  $\sigma_{\parallel}(\mathbf{r}, \omega) = \lim_{k \rightarrow 0} \sigma_{\parallel}(\mathbf{r}, k, \omega)$  and  $\gamma(\mathbf{r}, \omega) = \lim_{k \rightarrow 0} \gamma(\mathbf{r}, k, \omega)$ , the cut-off function  $\theta(\mathbf{r}', \mathbf{r})$  has—due to the rapid oscillations of the exponential for large  $|\mathbf{r} - \mathbf{r}'|$ —no effect [with regard to an application of the operator associated with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  from Eq. (104)] and can be dropped, and we obtain, as a generalization of Eq. (77),

$$\begin{aligned} \vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}(\mathbf{r}', \omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad - \nabla \times [\gamma(\mathbf{r}', \omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}')] \times \vec{\nabla}'. \end{aligned} \quad (107)$$

With the identifications

$$\sigma_{\parallel}(\mathbf{r}, \omega) = \varepsilon_0 \omega \operatorname{Im} \varepsilon(\mathbf{r}, \omega), \quad (108)$$

$$\gamma(\mathbf{r}, \omega) = -\kappa_0 \operatorname{Im} \kappa(\mathbf{r}, \omega) / \omega \quad (109)$$

[cf. Eqs. (82) and (83)], Eqs. (78) and (79) generalize to

$$\begin{aligned} \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) &= -i\varepsilon_0 \omega [\varepsilon(\mathbf{r}', \omega) - 1] \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad - \frac{1}{i\omega} \nabla \times \{\kappa_0 [1 - \kappa(\mathbf{r}', \omega)] \vec{I} \delta(\mathbf{r} - \mathbf{r}')\} \times \vec{\nabla}' \end{aligned} \quad (110)$$

and

$$\begin{aligned} \underline{\mathbf{j}}(\mathbf{r}, \omega) &= -i\varepsilon_0 \omega [\varepsilon(\mathbf{r}, \omega) - 1] \underline{\hat{\mathbf{E}}}(\mathbf{r}, \omega) \\ &\quad + \kappa_0 \nabla \times \{[1 - \kappa(\mathbf{r}, \omega)] \underline{\hat{\mathbf{B}}}(\mathbf{r}, \omega)\} + \underline{\mathbf{j}}_N(\mathbf{r}, \omega), \end{aligned} \quad (111)$$

respectively.

Unfortunately, Eq. (84) does not generalize to

$$\begin{aligned} \vec{K}'(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}^{1/2}(\mathbf{r}', \omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad \mp i\gamma^{1/2}(\mathbf{r}', \omega) \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}') \quad (\text{wrong!}), \end{aligned} \quad (112)$$

as could have been suspected. Indeed, straightforward calculation shows that, for spatially varying permittivity and permeability, the kernel (112) does not solve Eq. (18) [with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  as given in Eq. (107)], which implies that  $\underline{\mathbf{j}}_N(\mathbf{r}, \omega)$  cannot be related to the variables  $\underline{\mathbf{f}}(\mathbf{r}, \omega)$  as in Eq. (85), with  $\varepsilon(\omega)$  and  $\kappa(\omega)$  being simply replaced with their inhomogeneous counterparts  $\varepsilon(\mathbf{r}, \omega)$  and  $\kappa(\mathbf{r}, \omega)$ , respectively. In order to obtain an explicit expression for the kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  required in Eq. (17), one has instead to return to Eq. (18) and solve it with  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$  from Eq. (107)—a problem that is, however, very difficult to solve in general. Although this does not at all limit the practical applicability of the theory [since all one typically has to know about  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  is that it satisfies its defining equation (18) for the chosen conductivity (110)], it may be useful to have at hand, at least an approximate form for weak inhomogeneity, such as (App. D)

$$\begin{aligned} \vec{K}(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}^{1/2}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \mp i\gamma^{1/2}(\omega) \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad + \frac{1}{2} [\sigma_{\parallel}(\mathbf{r}, \omega) - \sigma_{\parallel}(\omega)] \vec{M}_0(\mathbf{r}, \mathbf{r}', \omega) \\ &\quad + \frac{1}{2} \nabla \times \{[\gamma(\mathbf{r}, \omega) - \gamma(\omega)] \nabla \times \vec{M}_0(\mathbf{r}, \mathbf{r}', \omega)\}, \end{aligned} \quad (113)$$

where

$$\begin{aligned} \vec{M}_0(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_{\parallel}^{-1/2}(\omega) \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &\quad \pm i\gamma^{-1/2}(\omega) \nabla \times m_0(\mathbf{r}, \mathbf{r}', \omega) \vec{I} \\ &\quad + \sigma_{\parallel}^{-1/2}(\omega) \nabla \times m_0(\mathbf{r}, \mathbf{r}', \omega) \vec{I} \times \vec{\nabla}', \end{aligned} \quad (114)$$

with  $m_0(\mathbf{r}, \mathbf{r}', \omega) = -(4\pi|\mathbf{r} - \mathbf{r}'|)^{-1} e^{-|\mathbf{r} - \mathbf{r}'|/\alpha(\omega)}$ ,  $\alpha(\omega) = [\gamma(\omega)/\sigma_{\parallel}(\omega)]^{1/2} > 0$ . If the lack of exact knowledge of  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  really happens to be an obstacle in an application, one can alternatively resort to the approach on the basis of Eqs. (89)–(91), by simply replacing therein  $\varepsilon(\omega)$  and  $\kappa(\omega)$  by  $\varepsilon(\mathbf{r}, \omega)$  and  $\kappa(\mathbf{r}, \omega)$ , respectively.

## V. GREEN TENSOR CONSTRUCTION FOR SPATIALLY DISPERSIVE BODIES

Practical application of the quantization scheme requires the solution of the classical problem of the determination of the Green tensor  $\vec{G}(\mathbf{r}, \mathbf{r}', \omega)$  for a given conductivity tensor  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$ . Typically, one has to deal with systems of bodies each of which can be regarded as being homogeneous in its respective interior region. The physical surfaces of the bodies, including the boundary

surfaces between adjacent bodies, may be said to be the particular space regions where the material properties differ significantly from the intra-body (bulk-material) properties. Physical surfaces are therefore not mathematical ones but more or less fuzzy boundary layers. For not too small bodies, however, they usually contain only a small fraction of the overall material, so that they may often be approximately replaced with (sharp) mathematical surfaces, with the idealization that the intra-body bulk-material properties hold immediately beyond them.

### A. Dielectric approximation

The application of the point of view just outlined to (systems of internally homogeneous) spatially dispersive bodies is commonly referred to as the dielectric approximation, for which it is obviously required that the characteristic length scale of spatial dispersion is small in comparison with the typical linear extensions of the bodies. Hence, making use of the same notation as employed in Eq. (100), we assume the conductivity tensor of a system of spatially dispersing bodies, in the dielectric approximation, to be of the form

$$\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) = \theta[\mathbf{L}(\mathbf{r}'), \mathbf{r}] \vec{Q}[\mathbf{L}(\mathbf{r}'), \mathbf{r} - \mathbf{r}', \omega], \quad (115)$$

with the vector  $\mathbf{L}$  labeling now the bodies in place of the lattice cells in Sec. IV C 1;  $\mathbf{L}(\mathbf{r}')$  singles out the particular body that contains the position  $\mathbf{r}'$ , and the quantities  $\vec{Q}[\mathbf{L}, \mathbf{r} - \mathbf{r}', \omega]$  are the bulk-material conductivity tensors ascribed to the various bodies. (Regions outside all actual bodies, if any, are formally viewed as bodies in this notation.) Note that  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  as given by Eq. (115)

satisfies the reciprocity condition. Corrections to the dielectric approximation in the form of surface currents or, equivalently, boundary conditions might be required for systems such as needle-shaped bodies, thin films or the like, where the dielectric approximation can be insufficient. However, as such corrections cannot be convincingly justified without detailed (model) assumptions about the specific nature of the physical surfaces, we do not consider them in the following.

### B. Integral equations and surface impedance method

The dielectric approximation renders it possible to formulate integral equations from which the Green tensor may be then derived. Here we outline the surface impedance method, which, in connection with its application to the calculation of Casimir forces, has recently given rise to controversial discussions (see, e.g., Refs. [24–27]) concerning the range of validity and the approximations involved in this method.

To begin with, let us consider spatially dispersive material described by a conductivity tensor  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  [not yet approximated by a form like Eq. (115)], and let  $V$  denote some space region of interest. Further, let  $\vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  be the Green tensor for an auxiliary problem to be specified yet. Then, if  $\mathbf{j}(\mathbf{r}, \omega)$  is an arbitrarily chosen current density in the frequency domain, given inside and/or outside  $V$ , and if  $\underline{\mathbf{E}}(\mathbf{r}, \omega)$  and  $\underline{\mathbf{B}}(\mathbf{r}, \omega) = (i\omega)^{-1} \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega)$ , respectively, are the (classical) electric and induction fields associated with this current density, the identity

$$\begin{aligned} & \int_V d^3r \underline{\mathbf{E}}(\mathbf{r}, \omega) \cdot \nabla \times \vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) + i\varepsilon_0\omega \int_V d^3r \underline{\mathbf{E}}(\mathbf{r}, \omega) \cdot \left[ \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) - (i\varepsilon_0\omega)^{-1} \int_V d^3s \vec{Q}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{s}, \mathbf{r}', \omega) \right] \\ &= \int_V d^3r \underline{\mathbf{j}}_V(\mathbf{r}, \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) + \int_{\partial V} da(\mathbf{r}) [\underline{\mathbf{E}}(\mathbf{r}, \omega) \times \mathbf{e}_n(\mathbf{r})] \cdot \vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) \\ & \quad + \mu_0^{-1} \int_{\partial V} da(\mathbf{r}) [\underline{\mathbf{B}}(\mathbf{r}, \omega) \times \mathbf{e}_n(\mathbf{r})] \cdot \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) \quad (116) \end{aligned}$$

(being a Green-type formula) holds, as can be proven correct by partial integration and employing the reciprocity property of  $\vec{Q}(\mathbf{r}, \mathbf{s}, \omega)$ . Here,  $da(\mathbf{r})$  and  $\mathbf{e}_n(\mathbf{r})$  denote the absolute value and the unit vector of the surface element

at  $\mathbf{r}$  on the surface  $\partial V$  of  $V$ . Further,

$$\begin{aligned} \underline{\mathbf{j}}_V(\mathbf{r}, \omega) = & (i\mu_0\omega)^{-1} \left[ \nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) \right. \\ & \left. - \frac{\omega^2}{c^2} \underline{\mathbf{E}}(\mathbf{r}, \omega) - i\mu_0\omega \int_V d^3r' \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \underline{\mathbf{E}}(\mathbf{r}', \omega) \right] \quad (117) \end{aligned}$$

and

$$\vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) = (i\mu_0\omega)^{-1} \nabla \times \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega). \quad (118)$$

Note that  $\vec{\mathbf{j}}_V(\mathbf{r}, \omega)$  does not agree with  $\vec{\mathbf{j}}(\mathbf{r}, \omega)$ , in general, as the  $\mathbf{r}'$ -integral in Eq. (117) extends only over  $V$ .

Let us now assume that  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  can be treated in the dielectric approximation according to Eq. (115), at least for the medium in  $V$ . In this case,  $\vec{\mathbf{j}}_V(\mathbf{r}, \omega)$  agrees with  $\vec{\mathbf{j}}(\mathbf{r}, \omega)$  inside  $V$ , and  $\vec{Q}(\mathbf{r}, \mathbf{s}, \omega)$  in Eq. (116) can be replaced with the bulk-medium conductivity tensor  $\vec{Q}(\mathbf{L}_V, \mathbf{r} - \mathbf{s}, \omega)$  attributed to the medium in  $V$  ( $\mathbf{L}_V$  is the vector labeling region  $V$ ). Hence, if  $\vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  is required, for  $\mathbf{r} \in V$  (at least), to obey the equation

$$\begin{aligned} \nabla \times \nabla \times \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) \\ - i\mu_0\omega \int_V d^3s \vec{Q}(\mathbf{L}_V, \mathbf{r} - \mathbf{s}, \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{s}, \mathbf{r}', \omega) \\ = \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \end{aligned} \quad (119)$$

then Eq. (116) simplifies to

$$\begin{aligned} (i\mu_0\omega)^{-1} \underline{\mathbf{E}}(\mathbf{r}', \omega) \theta(\mathbf{L}_V, \mathbf{r}') = (i\mu_0\omega)^{-1} \underline{\mathbf{E}}^{(\text{in})}(\mathbf{r}', \omega) \\ + \int_{\partial V} da(\mathbf{r}) [\underline{\mathbf{E}}(\mathbf{r}, \omega) \times \mathbf{e}_n(\mathbf{r})] \cdot \vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) \\ + \mu_0^{-1} \int_{\partial V} da(\mathbf{r}) [\underline{\mathbf{B}}(\mathbf{r}, \omega) \times \mathbf{e}_n(\mathbf{r})] \cdot \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega), \end{aligned} \quad (120)$$

where

$$\begin{aligned} \underline{\mathbf{E}}^{(\text{in})}(\mathbf{r}, \omega) = i\mu_0\omega \int_V d^3r' \vec{\mathbf{j}}(\mathbf{r}', \omega) \cdot \vec{G}_{\text{aux}}(\mathbf{r}', \mathbf{r}, \omega) \\ = i\mu_0\omega \int_V d^3r' \vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \vec{\mathbf{j}}(\mathbf{r}', \omega). \end{aligned} \quad (121)$$

Since the characteristic length of spatial dispersion is assumed to be sufficiently small as compared with the linear extensions of  $V$ , we may extend, with little error, the  $s$ -integral in Eq. (119) to the whole space and, therefore, we may identify  $\vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  in Eq. (120) with the corresponding bulk-medium Green tensor as given in App. E. Note that Eq. (120) may be viewed as a statement of Huyghens' principle (for  $\mathbf{r}'$  inside  $V$ ) and of the extinction theorem (for  $\mathbf{r}'$  outside  $V$ ), see Refs. [23, 28], which have been well known as a suitable starting point for field calculations on the basis of integral equation methods, see, in particular, Refs. [3–5] where the Wiener–Hopf technique has been used to construct solutions for a particular functional form of  $\vec{Q}(\mathbf{L}_V, \mathbf{r} - \mathbf{s}, \omega)$ .

The method of surface impedance (see, e.g., Ref. [29]) consists in the assumption that a linear relation between the tangential field components of  $\underline{\mathbf{E}}(\mathbf{r}, \omega)$  and  $\underline{\mathbf{B}}(\mathbf{r}, \omega)$

exists on the surface  $\partial V$ ,

$$\begin{aligned} \mathbf{e}_n(\mathbf{r}) \times \underline{\mathbf{E}}(\mathbf{r}, \omega) \\ = \mu_0^{-1} \int_{\partial V} da(\mathbf{r}') [\vec{Z}(\mathbf{r}, \mathbf{r}', \omega) \times \mathbf{e}_n(\mathbf{r}')] \cdot [\underline{\mathbf{B}}(\mathbf{r}', \omega) \times \mathbf{e}_n(\mathbf{r}')] \end{aligned} \quad (122)$$

( $\mathbf{r}$  on  $\partial V$ ), where the tensor  $\vec{Z}(\mathbf{r}, \mathbf{r}', \omega)$  is the dyadic surface impedance. Equivalently, one may consider the inverted form

$$\begin{aligned} \mu_0^{-1} \underline{\mathbf{B}}(\mathbf{r}, \omega) \times \mathbf{e}_n(\mathbf{r}) \\ = \int_{\partial V} da(\mathbf{r}') [\underline{\mathbf{E}}(\mathbf{r}', \omega) \times \mathbf{e}_n(\mathbf{r}')] \cdot \vec{Y}(\mathbf{r}', \mathbf{r}, \omega) \cdot \vec{I}_n(\mathbf{r}), \end{aligned} \quad (123)$$

where  $\vec{I}_n(\mathbf{r}) = \vec{I} - \mathbf{e}_n(\mathbf{r})\mathbf{e}_n(\mathbf{r})$  is a tangential projector, and  $\vec{Y}(\mathbf{r}, \mathbf{r}', \omega)$  may be referred to as the surface admittance. Once the relation (122) [or (123)] has been adopted, one may convert Eq. (120) (with  $\mathbf{r}' \in V$ ) into an integral equation for  $\underline{\mathbf{B}}(\mathbf{r}, \omega)$  [or  $\underline{\mathbf{E}}(\mathbf{r}, \omega)$ ] inside  $V$ , which can then be solved in terms of  $\underline{\mathbf{E}}^{(\text{in})}(\mathbf{r}, \omega)$  and the surface impedance, without specifying the medium properties outside  $V$ .

At first glance, the method has some advantageous features so that it has been enjoying a reputation in the literature. One of these features is that the necessity to know the medium properties everywhere outside  $V$  is replaced, so to speak, by the necessity to know the surface impedance for  $\partial V$ , which is a great reduction at first glance. Another one is that the application of continuity conditions may be sidestepped to some extent. Both of these points are to be qualified, however, and must be seen in the context of the following remarks.

First, since the current density  $\vec{\mathbf{j}}(\mathbf{r}, \omega)$  outside  $V$  does not contribute to Eq. (121), the solution for  $\underline{\mathbf{E}}(\mathbf{r}, \omega)$  can be unique only if  $\vec{\mathbf{j}}(\mathbf{r}, \omega)$  is located completely inside  $V$ ; solutions to the homogeneous integral equation can then be excluded from consideration. The surface impedance method applied to the volume  $V$  can thus yield, for given surface impedance, the Green tensor  $\vec{G}(\mathbf{r}, \mathbf{r}', \omega)$  only for both  $\mathbf{r}$  and  $\mathbf{r}'$  located in  $V$  (which may be sufficient for many applications, e.g., in the calculation of dispersion forces). Second, Eqs. (122) and (123) implicitly demand that the tangential components of the electric and the induction field uniquely exist on the surface  $\partial V$  (i.e., they should be continuous across the surface), or else ambiguities were encountered. Quasi-local approximations of the conductivity tensor should hence be excluded if they contain magnetic-like singular terms [as the second one in Eq. (110)].

Provided that Eq. (122) [or (123)] holds, the method requires knowledge of  $\vec{Z}(\mathbf{r}, \mathbf{r}', \omega)$  [or  $\vec{Y}(\mathbf{r}, \mathbf{r}', \omega)$ ], which plays the role of an external input. If  $\vec{Z}(\mathbf{r}, \mathbf{r}', \omega)$  [or  $\vec{Y}(\mathbf{r}, \mathbf{r}', \omega)$ ] is not known from the very beginning, one can

try to determine it *a posteriori* from the solution found, by appropriately specifying the effect of the medium on the electromagnetic outside the space region under consideration as well. We proceed with an example where this line can be pursued explicitly, on the basis of the dielectric approximation.

### C. Example: Non-magnetic, planar systems

Let  $V$  denote the slab-like region between two parallel planes  $z=0$  and  $z=d$ , and let  $\mathbf{j}(\mathbf{r}, \omega)$  be located completely inside  $V$ . In order to apply the surface impedance method to the region  $V$ , we assume that the medium in  $V$  is non-magnetic and can be treated in the dielectric approximation. Introducing Fourier transforms according to  $[\mathbf{r} \equiv (\boldsymbol{\rho}, z)]$

$$\underline{\mathbf{E}}(\mathbf{r}, \omega) = \int d^2q e^{i\mathbf{q} \cdot \boldsymbol{\rho}} \underline{\mathbf{E}}(z, \mathbf{q}, \omega) \quad (124)$$

and taking into account the lateral translational invariance of the system, we may write Eq. (123) in the Fourier domain as

$$\begin{aligned} & \mu_0^{-1} \underline{\mathbf{B}}(z, \mathbf{q}, \omega) \times \mathbf{e}_z \\ &= (2\pi)^2 \{ [\underline{\mathbf{E}}(d, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{Y}(d, z, -\mathbf{q}, \omega) \\ & \quad - [\underline{\mathbf{E}}(0, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{Y}(0, z, -\mathbf{q}, \omega) \} \cdot \vec{I}_z \end{aligned} \quad (125)$$

$[\vec{I}_z = \vec{I} - \mathbf{e}_z \mathbf{e}_z]$ , where  $z$  takes on the two values  $z=0$  and  $z=d$  (the two terms enter with opposite signs because of the opposite surface normals on the two sides of  $\partial V$ ). Similarly, the Fourier transformed version of Eq. (120) reads

$$\begin{aligned} & \underline{\mathbf{E}}(z', \mathbf{q}, \omega) \theta(\mathbf{L}_V, z') - \underline{\mathbf{E}}^{(\text{in})}(z', \mathbf{q}, \omega) \\ &= i\mu_0\omega(2\pi)^2 \{ [\underline{\mathbf{E}}(d, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{\Gamma}_{\text{aux}}(d, z', -\mathbf{q}, \omega) \\ & \quad - [\underline{\mathbf{E}}(0, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{\Gamma}_{\text{aux}}(0, z', -\mathbf{q}, \omega) \\ & \quad + \mu_0^{-1} [\underline{\mathbf{B}}(d, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{G}_{\text{aux}}(d, z', -\mathbf{q}, \omega) \\ & \quad - \mu_0^{-1} [\underline{\mathbf{B}}(0, \mathbf{q}, \omega) \times \mathbf{e}_z] \cdot \vec{G}_{\text{aux}}(0, z', -\mathbf{q}, \omega) \}. \end{aligned} \quad (126)$$

Note that  $\vec{G}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  and  $\vec{\Gamma}_{\text{aux}}(\mathbf{r}, \mathbf{r}', \omega)$  are translationally invariant, because they refer to bulk material. By means of Eq. (125), Eq. (126) takes the form

$$\begin{aligned} & \underline{\mathbf{E}}(z', \mathbf{q}, \omega) \theta(\mathbf{L}_V, z') - \underline{\mathbf{E}}^{(\text{in})}(z', \mathbf{q}, \omega) \\ &= \underline{\mathbf{E}}(d, \mathbf{q}, \omega) \cdot \vec{I}_z \cdot \vec{R}(d, z', \mathbf{q}, \omega) \\ & \quad - \underline{\mathbf{E}}(0, \mathbf{q}, \omega) \cdot \vec{I}_z \cdot \vec{R}(0, z', \mathbf{q}, \omega), \end{aligned} \quad (127)$$

with  $(\zeta=0, \zeta=d)$

$$\begin{aligned} \vec{R}(\zeta, z', \mathbf{q}, \omega) &= i\mu_0\omega(2\pi)^2 \mathbf{e}_z \times \left\{ \vec{\Gamma}_1(\zeta, z', -\mathbf{q}, \omega) \right. \\ & \quad + (2\pi)^2 \vec{Y}(\zeta, d, -\mathbf{q}, \omega) \cdot \vec{I}_z \cdot \vec{G}_1(d, z', -\mathbf{q}, \omega) \\ & \quad \left. - (2\pi)^2 \vec{Y}(\zeta, 0, -\mathbf{q}, \omega) \cdot \vec{I}_z \cdot \vec{G}_1(0, z', -\mathbf{q}, \omega) \right\} \end{aligned} \quad (128)$$

[note that  $\mathbf{e}_z \cdot \vec{R}(\zeta, z', \mathbf{q}, \omega) = 0$ ].

From Eq. (127) it follows that (Appendix F)

$$\begin{aligned} \underline{\mathbf{E}}(0, \mathbf{q}, \omega) \cdot \vec{I}_z &= [\underline{\mathbf{E}}^{(\text{in})}(0+, \mathbf{q}, \omega) \cdot \vec{C}^\sharp \\ & \quad - \underline{\mathbf{E}}^{(\text{in})}(d-, \mathbf{q}, \omega) \cdot \vec{D}^\sharp] \cdot [\vec{A} \cdot \vec{C}^\sharp - \vec{B} \cdot \vec{D}^\sharp]^\sharp, \end{aligned} \quad (129)$$

$$\begin{aligned} \underline{\mathbf{E}}(d, \mathbf{q}, \omega) \cdot \vec{I}_z &= [\underline{\mathbf{E}}^{(\text{in})}(0+, \mathbf{q}, \omega) \cdot \vec{A}^\sharp \\ & \quad - \underline{\mathbf{E}}^{(\text{in})}(d-, \mathbf{q}, \omega) \cdot \vec{B}^\sharp] \cdot [\vec{C} \cdot \vec{A}^\sharp - \vec{D} \cdot \vec{B}^\sharp]^\sharp, \end{aligned} \quad (130)$$

where

$$\vec{A} = \vec{A}(\mathbf{q}, \omega) = \vec{I}_z \cdot [\vec{I} + \vec{R}(0, 0+, \mathbf{q}, \omega)] \cdot \vec{I}_z, \quad (131)$$

$$\vec{B} = \vec{B}(\mathbf{q}, \omega) = \vec{I}_z \cdot \vec{R}(0, d-, \mathbf{q}, \omega) \cdot \vec{I}_z, \quad (132)$$

$$\vec{C} = \vec{C}(\mathbf{q}, \omega) = -\vec{I}_z \cdot \vec{R}(d, 0+, \mathbf{q}, \omega) \cdot \vec{I}_z, \quad (133)$$

$$\vec{D} = \vec{D}(\mathbf{q}, \omega) = \vec{I}_z \cdot [\vec{I} - \vec{R}(d, d-, \mathbf{q}, \omega)] \cdot \vec{I}_z, \quad (134)$$

and the superscript  $\sharp$  denotes matrix inversion with respect to the  $\vec{I}_z$ -space:  $\vec{A}^\sharp = \vec{I}_z \cdot (\vec{I}_z \cdot \vec{A} \cdot \vec{I}_z)^{-1} \cdot \vec{I}_z$ . The solution to Eq. (127) can now be readily obtained by substituting Eqs. (129) and (130) back in the right-hand side of Eq. (127). Specifying in Eq. (121) the source of  $\underline{\mathbf{E}}^{(\text{in})}(\mathbf{r}, \omega)$  so as to correspond to a point dipole situated in  $V$ ,  $\mathbf{j}(\mathbf{r}, \omega) = (i\mu_0\omega)^{-1} \mathbf{p} \delta(\mathbf{r} - \mathbf{r}_p)$ , from the corresponding position-space solution  $\underline{\mathbf{E}}(\mathbf{r}, \omega) \theta(\mathbf{L}_V, \mathbf{r})$ , one can then read off, according to  $\underline{\mathbf{E}}(\mathbf{r}, \omega) \theta(\mathbf{L}_V, \mathbf{r}) = \mathbf{p} \cdot \vec{G}(\mathbf{r}_p, \mathbf{r}, \omega) \theta(\mathbf{L}_V, \mathbf{r}) \theta(\mathbf{L}_V, \mathbf{r}_p)$ , the (interesting part of the) Green tensor expressed in terms of the surface admittance, i.e., in terms of the quantities  $\vec{Y}(z, z', \mathbf{q}, \omega)$  ( $z=0, d; z'=0, d$ ).

In order to illustrate the calculation of the admittance, let us assume that the (non-magnetic) media to the left and right of  $z=0$  and  $z=d$ , respectively, are (in the sense of the dielectric approximation) homogeneous half-spaces. Using the indices 0, 1, and 2 to distinguish the left ( $z < 0$ ) half-space, the volume  $V$  ( $0 < z < d$ ), and the right half-space ( $z > d$ ), respectively, and associating with each of the three regions the corresponding bulk-medium Green tensor  $\vec{G}_j(\mathbf{r}, \mathbf{r}', \omega)$  (App. E) and the associated auxiliary tensor  $\vec{\Gamma}_j(\mathbf{r}, \mathbf{r}', \omega)$  defined according to Eq. (118), one can derive the admittance by applying the

continuity conditions. Introducing the abbreviations

$$\vec{M}(\mathbf{q}, \omega) = -\vec{I}_z \cdot \vec{G}_1(0, d-, \mathbf{q}, \omega) \cdot \vec{I}_z, \quad (135)$$

$$\vec{N}(\mathbf{q}, \omega) = -\vec{I}_z \cdot [\vec{G}_1(0, 0+, \mathbf{q}, \omega) + \vec{G}_0(0, 0-, \mathbf{q}, \omega)] \cdot \vec{I}_z, \quad (136)$$

$$\vec{P}(\mathbf{q}, \omega) = \vec{I}_z \cdot [\vec{G}_1(d, d-, \mathbf{q}, \omega) + \vec{G}_2(0, 0+, \mathbf{q}, \omega)] \cdot \vec{I}_z, \quad (137)$$

$$\vec{S}(\mathbf{q}, \omega) = \vec{I}_z \cdot \vec{G}_1(d, 0+, \mathbf{q}, \omega) \cdot \vec{I}_z, \quad (138)$$

$$\vec{T}(\mathbf{q}, \omega) = \vec{I}_z \cdot \vec{\Gamma}_1(0, d-, \mathbf{q}, \omega) \cdot \vec{I}_z, \quad (139)$$

$$\vec{U}(\mathbf{q}, \omega) = \vec{I}_z \cdot [\vec{\Gamma}_1(0, 0+, \mathbf{q}, \omega) + \vec{\Gamma}_0(0, 0-, \mathbf{q}, \omega)] \cdot \vec{I}_z, \quad (140)$$

$$\vec{V}(\mathbf{q}, \omega) = -\vec{I}_z \cdot [\vec{\Gamma}_1(d, d-, \mathbf{q}, \omega) + \vec{\Gamma}_2(0, 0+, \mathbf{q}, \omega)] \cdot \vec{I}_z, \quad (141)$$

$$\vec{W}(\mathbf{q}, \omega) = -\vec{I}_z \cdot \vec{\Gamma}_1(d, 0+, \mathbf{q}, \omega) \cdot \vec{I}_z, \quad (142)$$

one eventually finds (App. G) the  $\vec{Y}(z, z', \mathbf{q}, \omega)$  ( $z=0, d$ ;  $z'=0, d$ ) to be (for notational convenience, the arguments  $\mathbf{q}, \omega$  are suppressed)

$$\vec{Y}(0, 0) = \frac{1}{(2\pi)^2} [\vec{U} \cdot \vec{S}^\# - \vec{T} \cdot \vec{P}^\#] \cdot [\vec{M} \cdot \vec{P}^\# - \vec{N} \cdot \vec{S}^\#]^\#, \quad (143)$$

$$\vec{Y}(0, d) = \frac{1}{(2\pi)^2} [\vec{U} \cdot \vec{N}^\# - \vec{T} \cdot \vec{M}^\#] \cdot [\vec{P} \cdot \vec{M}^\# - \vec{S} \cdot \vec{N}^\#]^\#, \quad (144)$$

$$\vec{Y}(d, 0) = \frac{1}{(2\pi)^2} [\vec{V} \cdot \vec{P}^\# - \vec{W} \cdot \vec{S}^\#] \cdot [\vec{M} \cdot \vec{P}^\# - \vec{N} \cdot \vec{S}^\#]^\#, \quad (145)$$

$$\vec{Y}(d, d) = \frac{1}{(2\pi)^2} [\vec{V} \cdot \vec{M}^\# - \vec{W} \cdot \vec{N}^\#] \cdot [\vec{P} \cdot \vec{M}^\# - \vec{S} \cdot \vec{N}^\#]^\#, \quad (146)$$

which are, quite naturally, far more complicated than the simple Kliever–Fuchs expressions mentioned in App. E.

## VI. SUMMARY AND CONCLUDING REMARKS

We have developed a rather general quantization scheme for the macroscopic electromagnetic field in arbitrary linearly responding media, which offers a unified approach to QED in linear media. Describing the medium response by a non-local conductivity tensor, any of the possible electromagnetic features of a linear medium in equilibrium is covered by the scheme, in particular, spatial dispersion. Central quantities of the scheme are the noise current that is intimately connected with the absorption necessarily observed in any linear medium in

equilibrium, the bosonic dynamical variables associated with the noise current, and the Green tensor of the phenomenological Maxwell equations, in which the medium properties enter via the conductivity tensor.

From a careful analysis of the dynamical variables and (quasi-)local limiting forms of the non-local conductivity tensor, we have shown how quantization schemes previously developed for locally responding media can be recovered as special applications of the general quantization scheme. In particular, a locally responding magnetodielectric medium can be viewed as a special quasi-local limiting case of an isotropic, spatially dispersive medium without optical activity, where the (local) dielectric permittivity and magnetic permeability are just two contributions to one and the same quasi-local conductivity tensor. As a result, application of the general quantization scheme shows that the electromagnetic field in such a medium can be quantized by using a single set of bosonic variables.

Generally, the use of a single set of bosonic variables means that the noise current which enters the macroscopic Maxwell equations is not divided into parts (associated, e.g., with a polarization and a magnetization) regarded as representing independent degrees of freedom, but is rather treated as an entity. This may be particularly advantageous for future studies of (quantum) electrodynamics in moving media, simplifying the discussion of transformations to different frames of reference. However, the theory also admits, by appropriate projection, the use of several independent sets of bosonic variables, which in fact corresponds to the neglect of certain kinds of interactions in the sense of super-selection rules.

Since exact solutions of Maxwell's equations are not available in closed form in general, even more so if spatial dispersion is taken into account, one has to resort to approximation methods to obtain explicit expressions for the Green tensor, the latter being one of the cornerstones of the theory. To assist in such intentions we have considered in some detail the dielectric approximation for the conductivity tensor, which consists in approximating the conductivity tensor of a system of spatially dispersive bodies by joining together bulk-medium conductivity tensors (which are routinely handled in reciprocal space). Although the information relevant to physical surface regions is lost in this way, the dielectric approximation has been a key tool to render tractable electromagnetic propagation problems in spatially dispersive media. Accepting the dielectric approximation, the problem of finding the Green tensor becomes then solvable via integral-equation and surface-impedance techniques.

As already mentioned, diamagnetic media are not covered by the quantization scheme developed in this paper—a scheme that exhausts the possibilities offered by the linear-response framework. Furthermore, it should be pointed out that the scheme does also not automatically apply to linearly amplifying media. Although both types of media do not really fit into the linear-response framework, they may be forced into it, but not without

reservations and alterations of the whole scheme. Clearly, the concept of linear amplification has a range of applicability very much smaller than that of linear dissipation.

Concluding, this work provides the most general quantization scheme for the electromagnetic field in linearly responding, absorbing materials to date, from which previously given schemes can be recovered as limiting cases. It will serve as a foundation for investigations of surface plasmon effects involving strong spatial dispersion, and as a starting point for the investigation of moving media.

## APPENDIX A: DERIVATION OF EQ. (8)

The linear integro-differential equation (8) can be represented as

$$\int d^3s \vec{H}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{G}(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{A1})$$

where the integral kernel

$$\begin{aligned} \vec{H}(\mathbf{r}, \mathbf{r}', \omega) &= \nabla \times \nabla \times \vec{I} \delta(\mathbf{r} - \mathbf{r}') \\ &- \frac{\omega^2}{c^2} \vec{I} \delta(\mathbf{r} - \mathbf{r}') - i\mu_0\omega \vec{Q}(\mathbf{r}, \mathbf{r}', \omega) \end{aligned} \quad (\text{A2})$$

is reciprocal,

$$\vec{H}(\mathbf{r}, \mathbf{r}', \omega) = \vec{H}^\top(\mathbf{r}', \mathbf{r}, \omega), \quad (\text{A3})$$

since  $\vec{Q}(\mathbf{r}, \mathbf{r}', \omega)$  is reciprocal. Hence, the transposed equation of Eq. (A1) takes the form

$$\int d^3s \vec{G}^\top(\mathbf{s}, \mathbf{r}, \omega) \cdot \vec{H}(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A4})$$

Multiplying from the right with  $\vec{G}(\mathbf{r}', \mathbf{s}', \omega)$ , integrating over  $\mathbf{r}'$ , and using Eq. (A1), one can see that the Green tensor is also reciprocal,

$$\vec{G}(\mathbf{r}, \mathbf{r}', \omega) = \vec{G}^\top(\mathbf{r}', \mathbf{r}, \omega). \quad (\text{A5})$$

Because of Eq. (A5), the complex conjugate of Eq. (A4) reads

$$\int d^3s \vec{G}^*(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{H}^*(\mathbf{s}, \mathbf{r}', \omega) = \vec{I} \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A6})$$

Taking the dot product of Eq. (A1) from the left with  $\vec{G}^*(\mathbf{s}', \mathbf{r}, \omega)$  and integrating over  $\mathbf{r}$ , taking the dot product of Eq. (A6) from the right with  $\vec{G}(\mathbf{r}', \mathbf{s}')$  and integrating over  $\mathbf{r}'$ , and subtracting the two resulting equations, one derives

$$\begin{aligned} \text{Im } \vec{G}(\mathbf{r}, \mathbf{r}', \omega) &= \\ - \int d^3s \int d^3s' \vec{G}(\mathbf{r}, \mathbf{s}, \omega) \cdot [\text{Im } \vec{H}(\mathbf{s}, \mathbf{s}', \omega)] \cdot \vec{G}^*(\mathbf{s}', \mathbf{r}', \omega). \end{aligned} \quad (\text{A7})$$

From Eq. (A2) it is seen that

$$\begin{aligned} \text{Im } \vec{H}(\mathbf{r}, \mathbf{r}', \omega) &= \\ - \frac{\text{Im } \omega^2}{c^2} \vec{I} \delta(\mathbf{r} - \mathbf{r}') - \mu_0 \text{Re} [\omega \vec{Q}(\mathbf{r}, \mathbf{r}', \omega)]. \end{aligned} \quad (\text{A8})$$

Insertion of Eq. (A8) in Eq. (A7) and restriction to real frequencies leads, upon recalling Eq. (2), to Eq. (8).

## APPENDIX B: NON-UNIQUENESS OF THE KERNEL $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$

The transition from  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  to  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  according to Eq. (24) can be re-interpreted as a redefinition of the dynamical variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$  according to

$$\hat{\mathbf{f}}(\mathbf{r}, \omega) = \int d^3r' \vec{V}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}'(\mathbf{r}', \omega), \quad (\text{B1})$$

$$\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) = \int d^3r' \vec{V}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}'^\dagger(\mathbf{r}', \omega). \quad (\text{B2})$$

Inserting Eq. (B1) into Eq. (17) yields

$$\hat{\mathbf{j}}_N(\mathbf{r}, \omega) = \left( \frac{\hbar\omega}{\pi} \right)^{\frac{1}{2}} \int d^3r' \vec{K}'(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}'(\mathbf{r}', \omega), \quad (\text{B3})$$

where  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  is just given by Eq. (24). With regard to the transformation (B1) and (B2), the significance of replacing Eq. (25) with Eq. (26) is that the variables  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}'^\dagger(\mathbf{r}, \omega)$  are uniquely expressible in terms of the  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$ , and so are on an equal footing with them—the unitary operator associated with  $\vec{V}(\mathbf{r}, \mathbf{r}', \omega)$  uniquely maps a set of bosonic variables onto a fully equivalent set of bosonic variables. Hence,  $\vec{V}(\mathbf{r}, \mathbf{r}', \omega)$  may be thought of as being included in the chosen set of dynamical variables. In this sense, it is sufficient to base the calculations in Sec. III on the Hermitian operator associated with the integral kernel  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  as defined by Eq. (23).

It is worth noting that the operator associated with  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  as defined by Eq. (24) is non-Hermitian whenever  $\vec{V}(\mathbf{r}, \mathbf{r}', \omega)$  is non-trivial. To see this, let us conversely assume that the operator associated with  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  is Hermitian,

$$\begin{aligned} \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}(\mathbf{s}, \mathbf{r}', \omega) \\ = \int d^3s \vec{V}^+(\mathbf{s}, \mathbf{r}, \omega) \cdot \vec{K}(\mathbf{s}, \mathbf{r}', \omega). \end{aligned} \quad (\text{B4})$$

Applying from the left the operator associated with  $\vec{V}$  and from the right the operator associated with  $\vec{V}^+$  and



recalling Eq. (25), one sees that

$$\begin{aligned} \int d^3s \vec{V}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{K}(\mathbf{s}, \mathbf{r}', \omega) \\ = \int d^3s \vec{K}(\mathbf{r}, \mathbf{s}, \omega) \cdot \vec{V}^+(\mathbf{r}', \mathbf{s}, \omega). \end{aligned} \quad (\text{B5})$$

Applying the operator associated with  $\vec{K}$  from the left to Eq. (B4) and from the right to Eq. (B5) and comparing the results, one finds that the operators associated with  $\vec{V}$  and  $\vec{\sigma}$  commute [recall Eq. (18)], so that the operator associated with  $\vec{V}$  maps each (possibly degenerate) eigenspace of the operator associated with  $\vec{\sigma}$  onto itself. Specifically, this implies that the operator associated with  $\vec{V}$  commutes with the spectral projectors of the operator associated with  $\vec{\sigma}$  [and, therefore, also with the projectors (34)]. Since the spectral projectors of the operators associated with  $\vec{\sigma}$  and  $\vec{K}$  are the same [cf. Eqs. (22) and (23)], the operators associated with  $\vec{V}$  and  $\vec{K}$  also commute. But then, since the operator associated with  $\vec{K}$  is invertible (being a positive operator), Eq. (B4) [or Eq. (B5)] shows that the operator associated with  $\vec{V}$  is Hermitian, i.e.,

$$\vec{V}(\mathbf{r}, \mathbf{r}', \omega) = \vec{V}^+(\mathbf{r}', \mathbf{r}, \omega). \quad (\text{B6})$$

Since the operator associated with  $\vec{V}$  is also unitary, in the diagonal expansion

$$\vec{V}(\mathbf{r}, \mathbf{r}', \omega) = \int d\alpha v(\alpha, \omega) \mathbf{F}(\alpha, \mathbf{r}, \omega) \mathbf{F}^*(\alpha, \mathbf{r}', \omega), \quad (\text{B7})$$

one must have  $v(\alpha, \omega) = \pm 1$  for each  $\alpha$ , which means that the Hermitian operator associated with  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$  can differ from the operator associated with  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  only by the trivial type of unitary transformation that merely replaces some of the basis functions  $\mathbf{F}(\alpha, \mathbf{r}, \omega)$  with  $-\mathbf{F}(\alpha, \mathbf{r}, \omega)$ . Conversely, this shows that any (in this sense) non-trivial  $\vec{V}(\mathbf{r}, \mathbf{r}', \omega)$  necessarily yields a non-Hermitian  $\vec{K}'(\mathbf{r}, \mathbf{r}', \omega)$ .

### APPENDIX C: REDUCED STATE SPACE AND SUPER-SELECTION RULE

Let us consider the state space spanned by the Fock states associated with  $\hat{\mathbf{f}}(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$  so that an arbitrary, normalizable state  $|\phi\rangle$  in this space can be represented in the form

resented in the form

$$\begin{aligned} |\phi\rangle &= |0\rangle\langle 0|\phi\rangle \\ &+ \sum_{k_1=1}^3 \int_0^\infty d\omega_1 \int d^3r_1 \phi_{k_1}(\mathbf{r}_1, \omega_1) |1_{k_1}(\mathbf{r}_1, \omega_1)\rangle \\ &+ \sum_{k_1, k_2=1}^3 \int_0^\infty d\omega_1 \int_0^\infty d\omega_2 \int d^3r_1 \int d^3r_2 \\ &\times \phi_{k_1 k_2}(\mathbf{r}_1, \omega_1, \mathbf{r}_2, \omega_2) |1_{k_1}(\mathbf{r}_1, \omega_1), 1_{k_2}(\mathbf{r}_2, \omega_2)\rangle \\ &+ \dots, \end{aligned} \quad (\text{C1})$$

where

$$\hat{f}_k(\mathbf{r}, \omega) |0\rangle = 0, \quad (\text{C2})$$

$$\hat{f}_k^\dagger(\mathbf{r}, \omega) |0\rangle = |1_k(\mathbf{r}, \omega)\rangle, \quad (\text{C3})$$

$$\begin{aligned} \hat{f}_{k_N}^\dagger(\mathbf{r}_N, \omega_N) \dots \hat{f}_{k_1}^\dagger(\mathbf{r}_1, \omega_1) |0\rangle \\ = |1_{k_1}(\mathbf{r}_1, \omega_1), \dots, 1_{k_N}(\mathbf{r}_N, \omega_N)\rangle. \end{aligned} \quad (\text{C4})$$

The normalization of  $|\phi\rangle$  can be obtained by using the formula (which can be viewed as a special case of the Bloch–De Dominicis theorem [14])

$$\begin{aligned} \langle 0 | \hat{f}_{k_M}(\mathbf{r}_M, \omega_M) \dots \hat{f}_{k_1}(\mathbf{r}_1, \omega_1) \\ \times \hat{f}_{k'_1}^\dagger(\mathbf{r}'_1, \omega'_1) \dots \hat{f}_{k'_N}^\dagger(\mathbf{r}'_N, \omega'_N) |0\rangle \\ = \delta_{MN} \sum_{\pi \in \mathcal{S}_N} \prod_{l=1}^N \delta_{k_l, k'_{\pi(l)}} \delta(\mathbf{r}_l - \mathbf{r}'_{\pi(l)}) \delta(\omega_l - \omega'_{\pi(l)}) \end{aligned} \quad (\text{C5})$$

( $\langle 0|0\rangle = 1$ ;  $\mathcal{S}_N$ , group of permutations of  $N$  objects).

In order to construct a reduced state space in which the operators  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  and  $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r}', \omega')$  defined by Eq. (35) behave like bosonic operators, let us first introduce states  $|0\rangle_\lambda$  according to

$$\hat{f}_{\lambda i}(\mathbf{r}, \omega) |0\rangle_\lambda = 0, \quad (\text{C6})$$

$$\hat{f}_{\lambda i}(\mathbf{r}, \omega) |0\rangle_{\lambda'} = |0\rangle_{\lambda'} \hat{f}_{\lambda i}(\mathbf{r}, \omega) \quad (\lambda \neq \lambda') \quad (\text{C7})$$

( $\lambda \langle 0|0\rangle_\lambda = 1$ ), such that

$$|0\rangle = \bigotimes_{\lambda=1}^{\Lambda} |0\rangle_\lambda. \quad (\text{C8})$$

Now let us introduce, for each  $\lambda$ , an orthogonal projector  $\hat{P}_\lambda$  as the sum of orthogonal projectors  $\hat{P}_\lambda^{(N)}$ ,

$$\hat{P}_\lambda = \sum_{N=0}^{\infty} \hat{P}_\lambda^{(N)}, \quad (\text{C9})$$

$$\hat{P}_\lambda^{(N)\dagger} = \hat{P}_\lambda^{(N)}, \quad (\text{C10})$$

$$\hat{P}_\lambda^{(N)} \hat{P}_{\lambda'}^{(N')} = \delta_{N N'} \hat{P}_\lambda^{(N)} \quad (\text{C11})$$

and specify  $\hat{P}_\lambda^{(N)}$  in such a way that, when applied to a quantum state of the form (C1), it picks out the

$(N+1)$ th term on the right-hand side of Eq. (C1) and incorporates  $N$  position-space projection kernels belonging to the chosen value of  $\lambda$ ,

$$\hat{P}_\lambda^{(0)} = |0\rangle_\lambda \langle 0|_\lambda \quad (\text{C12})$$

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$$\begin{aligned} \hat{P}_\lambda^{(N)} = & \frac{1}{N!} \sum_{k_1} \int_0^\infty d\omega_1 \int d^3 r_1 \sum_{k_2} \int_0^\infty d\omega_2 \int d^3 r_2 \cdots \sum_{k_N} \int_0^\infty d\omega_N \int d^3 r_N \\ & \times \hat{f}_{\lambda k_1}^\dagger(\mathbf{r}_1, \omega_1) \hat{f}_{\lambda k_2}^\dagger(\mathbf{r}_2, \omega_2) \cdots \hat{f}_{\lambda k_N}^\dagger(\mathbf{r}_N, \omega_N) \hat{P}_\lambda^{(0)} \hat{f}_{\lambda k_N}(\mathbf{r}_N, \omega_N) \hat{f}_{\lambda k_{N-1}}(\mathbf{r}_{N-1}, \omega_{N-1}) \cdots \hat{f}_{\lambda k_1}(\mathbf{r}_1, \omega_1) \end{aligned} \quad (\text{C13})$$


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$(N=1, 2, \dots)$ . It is not difficult to prove that Eqs. (C10) and (C11) are fulfilled, where the latter equation fixes the normalization factor  $1/N!$  in Eq. (C13), and that, in view of Eqs. (C8) and (C11), the commutation relation

$$[\hat{P}_\lambda^{(N)}, \hat{P}_{\lambda'}^{(N')}] = 0 \quad (\text{C14})$$

holds.

We may now define a reduced state space that contains only those (normalizable) vectors that have the separable form

$$|\phi\rangle^{(\text{red})} = \bigotimes_{\lambda=1}^{\Lambda} |\phi\rangle_\lambda, \quad (\text{C15})$$

$$\hat{P}_\lambda |\phi\rangle_\lambda = |\phi\rangle_\lambda, \quad (\text{C16})$$

with each vector  $|\phi\rangle_\lambda$  being, by construction, a superposition of vectors

$$\begin{aligned} |N\rangle_\lambda = & \sum_{k_1} \int_0^\infty d\omega_1 \int d^3 r_1 \cdots \sum_{k_N} \int_0^\infty d\omega_N \int d^3 r_N \\ & \times C_{\lambda k_1 \dots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N) \\ & \times |1_{\lambda k_1}(\mathbf{r}_1, \omega_1), \dots, 1_{\lambda k_N}(\mathbf{r}_N, \omega_N)\rangle, \end{aligned} \quad (\text{C17})$$

where, in analogy to Eq. (C4),

$$\begin{aligned} & |1_{\lambda k_1}(\mathbf{r}_1, \omega_1), \dots, 1_{\lambda k_N}(\mathbf{r}_N, \omega_N)\rangle \\ & = \hat{f}_{\lambda k_N}^\dagger(\mathbf{r}_N, \omega_N) \cdots \hat{f}_{\lambda k_1}^\dagger(\mathbf{r}_1, \omega_1) |0\rangle_\lambda. \end{aligned} \quad (\text{C18})$$

The important feature of these states is that the result of performing the integrations in Eq. (C17) is not changed if the wave function  $C_{\lambda k_1 \dots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)$  is replaced according to

$$\begin{aligned} & C_{\lambda k_1 \dots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N) \\ & \mapsto \int d^3 r'_1 \cdots \int d^3 r'_N (\vec{P}_\lambda)_{k_1 k'_1}(\mathbf{r}_1, \mathbf{r}'_1, \omega_1) \cdots \\ & \times (\vec{P}_\lambda)_{k_N k'_N}(\mathbf{r}_N, \mathbf{r}'_N, \omega_N) C_{\lambda k'_1 \dots \lambda k'_N}(\mathbf{r}'_1, \omega_1, \dots, \mathbf{r}'_N, \omega_N). \end{aligned} \quad (\text{C19})$$

It is also not changed if  $C_{\lambda k_1 \dots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N)$  is symmetrized with respect to the labels  $1, \dots, N$ . Wave functions that can be reduced to the same standardized wave function by these operations are thus fully equivalent representatives of the same vector. Without loss of generality, one can thus adopt the convention to employ only such standardized wave functions.

The commutation relation (36) implies that

$$\begin{aligned} & e^{\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)} \hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega') e^{-\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)} \\ & = \hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'} (\vec{P}_\lambda)_{k k'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega') \end{aligned} \quad (\text{C20})$$

with  $\epsilon$  being a parameter. As Eq. (C20) is a similarity transformation, it generalizes to

$$\begin{aligned} & e^{\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)} F[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')] e^{-\epsilon \hat{f}_{\lambda k}(\mathbf{r}, \omega)} \\ & = F[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega') + \epsilon \delta_{\lambda \lambda'} (\vec{P}_\lambda)_{k k'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')] \end{aligned} \quad (\text{C21})$$

where  $F = F[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')]$  is any well-behaved functional of  $\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')$ . Comparison of the terms of first order in  $\epsilon$  on both sides yields

$$\begin{aligned} & [\hat{f}_{\lambda k}(\mathbf{r}, \omega), F[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')]] \\ & = \left\{ \frac{\partial}{\partial \epsilon} F[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')] \right. \\ & \quad \left. + \epsilon \delta_{\lambda \lambda'} (\vec{P}_\lambda)_{k k'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega') \right\}_{\epsilon=0}. \end{aligned} \quad (\text{C22})$$

Let us consider the particular functional  $F_N[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')]$  appearing in Eqs. (C17), (C18),

$$\begin{aligned} & F_N[\hat{f}_{\lambda' k'}^\dagger(\mathbf{r}', \omega')] \\ & = \sum_{k_1} \int_0^\infty d\omega_1 \int d^3 r_1 \cdots \sum_{k_N} \int_0^\infty d\omega_N \int d^3 r_N \\ & \quad \times C_{\lambda k_1 \dots \lambda k_N}(\mathbf{r}_1, \omega_1, \dots, \mathbf{r}_N, \omega_N) \\ & \quad \times \hat{f}_{\lambda k_N}^\dagger(\mathbf{r}_N, \omega_N) \cdots \hat{f}_{\lambda k_1}^\dagger(\mathbf{r}_1, \omega_1). \end{aligned} \quad (\text{C23})$$

If the convention to use only standardized wave functions is adopted, one may write

$$\begin{aligned} F_N[\hat{f}_{\lambda'k'}^\dagger(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} (\vec{P}_\lambda)_{kk'}(\mathbf{r}, \mathbf{r}', \omega') \delta(\omega - \omega')] \\ = F_N[\hat{f}_{\lambda'k'}^\dagger(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} \delta_{kk'} \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega')], \end{aligned} \quad (\text{C24})$$

which means that the right-hand side of Eq. (C22) may be evaluated, for this functional, just as an ordinary functional derivative, i.e.,

$$\left[ \hat{f}_{\lambda k}(\mathbf{r}, \omega), F_N[\hat{f}_{\lambda'k'}^\dagger(\mathbf{r}', \omega')] \right] = \frac{\delta F_N[\hat{f}_{\lambda'k'}^\dagger(\mathbf{r}', \omega')]}{\delta \hat{f}_{\lambda k}^\dagger(\mathbf{r}, \omega)}. \quad (\text{C25})$$

But since, due to the definition of the reduced state space, only commutators of the type (C25) (for all  $N$ ) are required, and since Eq. (C25) can be obtained from Eq. (43) in the same way that Eq. (C22) has been obtained from Eq. (36), Eq. (43) is generally valid for the reduced state space.

#### APPENDIX D: DERIVATION OF EQ. (113)

For notational convenience, let us write here the integral equation (18) in the compact operator form

$$\mathcal{K}\mathcal{K}^\dagger = \sigma, \quad (\text{D1})$$

with  $\mathcal{K}$ , and  $\sigma$  being, respectively, the operators associated (for chosen  $\omega$ ) with the integral kernels  $\vec{K}(\mathbf{r}, \mathbf{r}', \omega)$  and  $\vec{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ . Accordingly, Eqs. (24) and (26) read  $\mathcal{K}' = \mathcal{K}\mathcal{V}$  and  $\mathcal{V}^\dagger\mathcal{V} = \mathcal{V}\mathcal{V}^\dagger = \mathcal{I}$ , respectively ( $\mathcal{I}$ , unit operator). Assuming that the (Hermitian and positive) operator  $\sigma$  takes the form

$$\sigma = \sigma_0 + \epsilon\sigma_1, \quad (\text{D2})$$

with  $\epsilon$  being a small, real parameter, we may try to find a solution to Eq. (D1) by the perturbative ansatz

$$\mathcal{K}' = \mathcal{K}'_0 + \epsilon\mathcal{K}'_1 + \dots, \quad (\text{D3})$$

where  $\mathcal{K}'_0$  is a solution to Eq. (D1) for  $\epsilon=0$ . Substituting Eqs. (D2) and (D3) into Eq. (D1), we see that the first-order correction  $\mathcal{K}'_1$  obeys the equation

$$\mathcal{K}'_0\mathcal{K}'_1^\dagger + \mathcal{K}'_1\mathcal{K}'_0^\dagger = \sigma_1, \quad (\text{D4})$$

which determines the Hermitian part of  $\mathcal{K}'_0\mathcal{K}'_1^\dagger$  (recall that  $\sigma_1$  is Hermitian), whereas the anti-Hermitian part is left undetermined. The solution to Eq. (D4) may therefore be written as

$$\mathcal{K}'_1 = \frac{1}{2}(\sigma_1 + \mathcal{A})\mathcal{M}_0, \quad (\text{D5})$$

where  $\mathcal{M}_0 = (\mathcal{K}'_0)^\dagger$ , and  $\mathcal{A} = -\mathcal{A}^\dagger$  is an arbitrary anti-Hermitian operator, which may be simply set to zero.

Note that the freedom to choose  $\mathcal{A}$  corresponds to the freedom to choose  $\mathcal{V}$ . We hence obtain to first order in  $\sigma - \sigma_0$

$$\mathcal{K}' = \mathcal{K}'_0 + \frac{1}{2}(\sigma - \sigma_0)\mathcal{M}_0. \quad (\text{D6})$$

If  $\sigma_0$ ,  $\mathcal{K}'_0$ , and  $\sigma$  are identified with the operators associated with the kernels (77), (84) and (107), respectively, Eq. (D6) is just the operator equivalent of Eq. (113)  $[\vec{K}'(\mathbf{r}, \mathbf{r}', \omega) \leftrightarrow \vec{K}(\mathbf{r}, \mathbf{r}', \omega)]$ .

To calculate explicitly the integral kernel  $\vec{M}_0(\mathbf{r}, \mathbf{r}', \omega)$  associated with  $\mathcal{M}_0$ , we consider the Fourier representation

$$\vec{K}'_0(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int d^3k \vec{K}'_0(\mathbf{k}, \omega) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \quad (\text{D7})$$

where, according to Eq. (71),

$$\vec{K}'_0(\mathbf{k}, \omega) = \sigma_\parallel^{1/2}(\omega) [\vec{I} \pm \alpha(\omega)\mathbf{k} \times \vec{I}] \quad (\text{D8})$$

$[\alpha(\omega) = [\gamma(\omega)/\sigma_\parallel(\omega)]^{1/2} > 0]$ , which shows that the kernel  $\vec{M}_0(\mathbf{r}, \mathbf{r}', \omega)$  can be given by the Fourier integral

$$\begin{aligned} \vec{M}_0(\mathbf{r}, \mathbf{r}', \omega) &= \sigma_\parallel^{-1/2}(\omega) \\ &\times \int \frac{d^3k}{(2\pi)^3} [\vec{I} \mp \alpha(\omega)\mathbf{k} \times \vec{I}]^{-1} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \end{aligned} \quad (\text{D9})$$

and it is not difficult to prove that

$$\begin{aligned} [\vec{I} \pm \alpha(\omega)\mathbf{k} \times \vec{I}]^{-1} &= \vec{I} \mp \alpha(\omega) \frac{\mathbf{k} \times \vec{I}}{1 + \alpha(\omega)^2 k^2} \\ &+ \alpha(\omega)^2 \frac{\mathbf{k} \times \vec{I} \times \mathbf{k}}{1 + \alpha(\omega)^2 k^2}. \end{aligned} \quad (\text{D10})$$

Introducing the function

$$\begin{aligned} m_0(\mathbf{r}, \mathbf{r}', \omega) &= \frac{1}{(2\pi)^3} \int d^3k \frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}}{\alpha(\omega)^{-2} + k^2} \\ &= -(4\pi|\mathbf{r} - \mathbf{r}'|)^{-1} e^{-|\mathbf{r}-\mathbf{r}'|/\alpha(\omega)}, \end{aligned} \quad (\text{D11})$$

we may rewrite Eq. (D9) [with Eq. (D10)] to obtain Eq. (114). Note that the (Yukawa-type) function  $m_0(\mathbf{r}, \mathbf{r}', \omega)$  satisfies the equation

$$[-\Delta + \alpha(\omega)^{-2}] m_0(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \quad (\text{D12})$$

together with the boundary condition  $m_0(\mathbf{r}, \mathbf{r}', \omega) \rightarrow 0$  for  $|\mathbf{r} - \mathbf{r}'| \rightarrow \infty$ .

#### APPENDIX E: BULK-MEDIUM GREEN TENSOR AND KLIEVER-FUCHS IMPEDANCE

For (translationally invariant) bulk material,

$$\vec{Q}(\mathbf{r}, \mathbf{r}', \omega) = \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \vec{Q}(\mathbf{k}, \omega), \quad (\text{E1})$$

the solution to Eq. (8) has the form

$$\vec{G}^{(0)}(\mathbf{r}, \mathbf{r}', \omega) = \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')} \vec{G}^{(0)}(\mathbf{k}, \omega), \quad (\text{E2})$$

where  $\vec{G}^{(0)}(\mathbf{k}, \omega)$  is the solution to a simple  $3 \times 3$  matrix equation. In particular, for an isotropic medium without optical activity,

$$\vec{Q}(\mathbf{k}, \omega) = Q_{\parallel}(k, \omega) \frac{\mathbf{k}\mathbf{k}}{k^2} + Q_{\perp}(k, \omega) \left( \vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right), \quad (\text{E3})$$

one finds that

$$\vec{G}^{(0)}(\mathbf{k}, \omega) = \frac{\vec{I} - \mathbf{k}\mathbf{k}/k^2}{D_{\perp}(k, \omega)} - \frac{\mathbf{k}\mathbf{k}/k^2}{D_{\parallel}(k, \omega)}, \quad (\text{E4})$$

where

$$D_{\perp}(k, \omega) = k^2 - \omega^2/c^2 - i\mu_0\omega Q_{\perp}(k, \omega), \quad (\text{E5})$$

$$D_{\parallel}(k, \omega) = \omega^2/c^2 + i\mu_0\omega Q_{\parallel}(k, \omega), \quad (\text{E6})$$

and

$$Q_{\parallel(\perp)}(k, \omega) = -i\varepsilon_0\omega[\varepsilon_{\parallel(\perp)}(k, \omega) - 1] \quad (\text{E7})$$

in ‘dielectric’ notation. In the general case

$$\begin{aligned} \vec{Q}(\mathbf{k}, \omega) &= Q_{\parallel}(\mathbf{k}, \omega) \frac{\mathbf{k}\mathbf{k}}{k^2} \\ &+ \left( \vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right) \cdot \vec{Q}_{\perp}(\mathbf{k}, \omega) \cdot \left( \vec{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right), \end{aligned} \quad (\text{E8})$$

$D_{\parallel}(k, \omega)$  changes to  $D_{\parallel}(\mathbf{k}, \omega)$ ,  $D_{\perp}(k, \omega)$  changes to the tensor

$$\vec{D}_{\perp}(\mathbf{k}, \omega) = k^2 - \omega^2/c^2 - i\mu_0\omega \vec{Q}_{\perp}(\mathbf{k}, \omega), \quad (\text{E9})$$

and the first term on the right-hand side of Eq. (E4) has to be replaced according to ( $\vec{I}_k = \vec{I} - \mathbf{k}\mathbf{k}/k^2$ )

$$\frac{\vec{I} - \mathbf{k}\mathbf{k}/k^2}{D_{\perp}(k, \omega)} \mapsto \vec{I}_k \cdot \left[ \vec{I}_k \cdot \vec{D}_{\perp}(\mathbf{k}, \omega) \cdot \vec{I}_k \right]^{-1} \cdot \vec{I}_k. \quad (\text{E10})$$

It may be convenient—particularly with regard to systems that are translationally invariant only in a plane, say the  $xy$  plane—to rewrite Eq. (E2) as  $[\mathbf{r} = (\boldsymbol{\rho}, z), \mathbf{k} = (\mathbf{q}, \beta)]$

$$\vec{G}^{(0)}(\mathbf{r}, \mathbf{r}', \omega) = \int d^2q e^{i\mathbf{q} \cdot (\boldsymbol{\rho} - \boldsymbol{\rho}')} \vec{G}^{(0)}(z, z', \mathbf{q}, \omega), \quad (\text{E11})$$

where

$$\vec{G}^{(0)}(z, z', \mathbf{q}, \omega) = \int \frac{d\beta}{(2\pi)^3} e^{i\beta(z-z')} \vec{G}^{(0)}(\mathbf{k}, \omega). \quad (\text{E12})$$

The analytical properties of the integrand in Eq. (E12) with respect to  $\beta$  depend on the specific decay to

zero of  $\vec{Q}(\mathbf{r} - \mathbf{r}', \omega)$  for  $|\mathbf{r} - \mathbf{r}'| \rightarrow \infty$ . For sufficiently rapid decay, Eq. (E12) admits an evaluation by contour integration in the complex  $\beta$  plane, which will be governed—focusing again on isotropic media without optical activity—by the solutions  $\beta = \beta_{\nu}^{\perp, \parallel}(q, \omega)$  of the two dispersion equations  $D_{\perp, \parallel}(k, \omega) = 0$ . In contrast to the case of spatially non-dispersive material, these equations are transcendental with respect to  $\beta$  rather than polynomial, so that nothing can be said about the number of their solutions in general. Specifically, if there are more than two functions  $\beta_{\nu}^{\perp}(q, \omega)$  [and/or one or more functions  $\beta_{\nu}^{\parallel}(q, \omega)$ ], the medium is said to support ‘additional’ (inhomogeneous plane-)waves.

We close this appendix with the following (perhaps not well-known) observation. Inserting Eq. (E4) in Eq. (E12) and setting  $z - z' = 0 \pm$ , and making use of the decompositions

$$\vec{I} - \mathbf{k}\mathbf{k}/k^2 = \mathbf{e}_s(\mathbf{q})\mathbf{e}_s(\mathbf{q}) + \mathbf{e}_p(\mathbf{k})\mathbf{e}_p(\mathbf{k}) \quad (\text{E13})$$

$$\vec{I}_z = \mathbf{e}_s(\mathbf{q})\mathbf{e}_s(\mathbf{q}) + \mathbf{q}\mathbf{q}/q^2, \quad (\text{E14})$$

with  $\mathbf{e}_s(\mathbf{q}) = \mathbf{q} \times \mathbf{e}_z/q$  and  $\mathbf{e}_p(\mathbf{k}) = \mathbf{k} \times \mathbf{q} \times \mathbf{e}_z/kq$  being polarization unit vectors, one can show that

$$\begin{aligned} \vec{I}_z \cdot \vec{G}^{(0)}(0 \pm, 0, \mathbf{q}, \omega) \cdot \vec{I}_z \\ = (i\mu_0\omega)^{-1} [Z_s(q, \omega) \mathbf{e}_s(\mathbf{q})\mathbf{e}_s(\mathbf{q}) + Z_p(q, \omega) \mathbf{q}\mathbf{q}/q^2], \end{aligned} \quad (\text{E15})$$

where

$$Z_s(q, \omega) = i\mu_0\omega \int_{-\infty}^{\infty} \frac{d\beta}{(2\pi)^3} \frac{e^{i\beta 0 \pm}}{D_{\perp}(k, \omega)}, \quad (\text{E16})$$

$$\begin{aligned} Z_p(q, \omega) &= i\mu_0\omega \\ &\times \int_{-\infty}^{\infty} \frac{d\beta}{(2\pi)^3} \frac{e^{i\beta 0 \pm}}{k^2} \left[ \frac{\beta^2}{D_{\perp}(k, \omega)} - \frac{q^2}{D_{\parallel}(k, \omega)} \right]. \end{aligned} \quad (\text{E17})$$

With Eqs. (E5)–(E7), Eqs. (E16) and (E17) are recognized (up to a trivial factor) as the surface impedance expressions first derived by Kliever and Fuchs [30] for a spatially dispersive half-space by assuming specular electron reflection (see also Ref. [27]).

## APPENDIX F: DERIVATION OF EQS. (129) AND (130)

From inspection of Eq. (127) it is seen that only the tangential components of the electric field, which are assumed to be continuous at  $z' = 0$  and  $z' = d$ , contribute to the right-hand side of this equation. We therefore evaluate Eq. (127) at  $z' = 0+$  and  $z' = d-$  and take the tangential ( $\vec{I}_z$ ) component thereof to obtain the linear equations (the arguments  $\mathbf{q}$  and  $\omega$  are kept fixed in this

appendix and are suppressed for notational convenience)

$$\begin{aligned} \underline{\mathbf{E}}(0+) \cdot \vec{I}_z \cdot [\vec{I} + \vec{R}(0, 0+)] \cdot \vec{I}_z \\ - \underline{\mathbf{E}}(d-) \cdot \vec{I}_z \cdot \vec{R}(d, 0+) \cdot \vec{I}_z = \underline{\mathbf{E}}^{(\text{in})}(0+) \cdot \vec{I}_z \end{aligned} \quad (\text{F1})$$

and

$$\begin{aligned} \underline{\mathbf{E}}(0+) \cdot \vec{I}_z \cdot \vec{R}(0, d-) \cdot \vec{I}_z \\ + \underline{\mathbf{E}}(d-) \cdot \vec{I}_z \cdot [\vec{I} - \vec{R}(d, d-)] \cdot \vec{I}_z = \underline{\mathbf{E}}^{(\text{in})}(d-) \cdot \vec{I}_z, \end{aligned} \quad (\text{F2})$$

respectively, which are to be solved for the tangential components  $\underline{\mathbf{E}}(0+) \cdot \vec{I}_z$  and  $\underline{\mathbf{E}}(d-) \cdot \vec{I}_z$ . To represent the solution in a compact form, we assign to any matrix  $\vec{A}$  satisfying  $\vec{I}_z \cdot \vec{A} \cdot \vec{I}_z = \vec{A}$  its inverse on the  $\vec{I}_z$ -space,  $\vec{A}^\# = \vec{I}_z \cdot (\vec{I}_z \cdot \vec{A} \cdot \vec{I}_z)^{-1} \cdot \vec{I}_z$ . It is straightforward to see that the block-matrix formula

$$\begin{pmatrix} \vec{A} & \vec{B} \\ \vec{C} & \vec{D} \end{pmatrix} \cdot \begin{pmatrix} \vec{C}^\# \cdot [\vec{A} \cdot \vec{C}^\# - \vec{B} \cdot \vec{D}^\#]^\# & \vec{A}^\# \cdot [\vec{C} \cdot \vec{A}^\# - \vec{D} \cdot \vec{B}^\#]^\# \\ \vec{D}^\# \cdot [\vec{B} \cdot \vec{D}^\# - \vec{A} \cdot \vec{C}^\#]^\# & \vec{B}^\# \cdot [\vec{D} \cdot \vec{B}^\# - \vec{C} \cdot \vec{A}^\#]^\# \end{pmatrix} = \begin{pmatrix} \vec{I}_z & 0 \\ 0 & \vec{I}_z \end{pmatrix} \quad (\text{F3})$$

is generally valid whenever the requisite inverse elements exist, so that the solution to Eqs. (F1) and (F2) can be written in the form

$$\begin{aligned} \underline{\mathbf{E}}(0+) \cdot \vec{I}_z = [\underline{\mathbf{E}}^{(\text{in})}(0+) \cdot \vec{C}^\# \\ - \underline{\mathbf{E}}^{(\text{in})}(d-) \cdot \vec{D}^\#] \cdot [\vec{A} \cdot \vec{C}^\# - \vec{B} \cdot \vec{D}^\#]^\#, \end{aligned} \quad (\text{F4})$$

$$\begin{aligned} \underline{\mathbf{E}}(d-) \cdot \vec{I}_z = [\underline{\mathbf{E}}^{(\text{in})}(0+) \cdot \vec{A}^\# \\ - \underline{\mathbf{E}}^{(\text{in})}(d-) \cdot \vec{B}^\#] \cdot [\vec{C} \cdot \vec{A}^\# - \vec{D} \cdot \vec{B}^\#]^\# \end{aligned} \quad (\text{F5})$$

together with Eqs. (131)–(134). Recalling again the continuity of the tangential component of the electric field, we are left with Eq. (129) and (130). It should be noted that the above inversion procedure fails at particular values of the (suppressed) arguments  $\mathbf{q}$  and  $\omega$ , because of singularities. However, as we are dealing with a lossy system, such singularities—corresponding to guided waves—may appear only when  $\text{Im } \omega < 0$  (for real values of  $\mathbf{q}$ ).

## APPENDIX G: DERIVATION OF Eqs. (143)–(146)

Let us attribute to the three regions  $j = 0$  ( $z < 0$ ),  $j = 1$  ( $0 < z < d$ ),  $j = 2$  ( $z > d$ ) bulk-medium conductivities  $\vec{Q}_j(\mathbf{r} - \mathbf{r}', \omega)$ , which combine to the overall conductivity tensor in the sense of Eq. (115) [ $j \leftrightarrow \mathbf{L}$ ]. For each of these regions, we construct, according to Eq. (118), the translationally invariant bulk-medium Green tensor  $\vec{G}_j(\mathbf{r}, \mathbf{r}', \omega)$  and the associated auxiliary tensor  $\vec{\Gamma}_j(\mathbf{r}, \mathbf{r}', \omega)$  in terms of their Fourier components  $\vec{G}_j(z, z', \mathbf{q}, \omega)$  and  $\vec{\Gamma}_j(z, z', \mathbf{q}, \omega)$  [defined according to Eqs. (E11), (E12)]. The three regions are thus described on an equal footing so that for the field in each region, an equation similar to Eq. (126) holds. Evaluating the tangential components of these three equations (which together determine the field in all of space) and using the continuity conditions at  $z = 0$  and  $z = d$ , one obtains two sets of equations for the tangential boundary values,

$$\begin{aligned} \{[\underline{\mathbf{E}}(d) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(d, d-) + [\mu_0^{-1} \underline{\mathbf{B}}(d) \times \mathbf{e}_z] \cdot \vec{G}_1(d, d-) - [\underline{\mathbf{E}}(0) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(0, d-) - [\mu_0^{-1} \underline{\mathbf{B}}(0) \times \mathbf{e}_z] \cdot \vec{G}_1(0, d-)\} \cdot \vec{I}_z \\ = -\{[\underline{\mathbf{E}}(d) \times \mathbf{e}_z] \cdot \vec{\Gamma}_2(0, 0+) + [\mu_0^{-1} \underline{\mathbf{B}}(d) \times \mathbf{e}_z] \cdot \vec{G}_2(0, 0+)\} \cdot \vec{I}_z \end{aligned} \quad (\text{G1})$$

and

$$\begin{aligned} \{[\underline{\mathbf{E}}(d) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(d, 0+) + [\mu_0^{-1} \underline{\mathbf{B}}(d) \times \mathbf{e}_z] \cdot \vec{G}_1(d, 0+) - [\underline{\mathbf{E}}(0) \times \mathbf{e}_z] \cdot \vec{\Gamma}_1(0, 0+) - [\mu_0^{-1} \underline{\mathbf{B}}(0) \times \mathbf{e}_z] \cdot \vec{G}_1(0, 0+)\} \cdot \vec{I}_z \\ = \{[\underline{\mathbf{E}}(0) \times \mathbf{e}_z] \cdot \vec{\Gamma}_0(0, 0-) + [\mu_0^{-1} \underline{\mathbf{B}}(0) \times \mathbf{e}_z] \cdot \vec{G}_0(0, 0-)\} \cdot \vec{I}_z, \end{aligned} \quad (\text{G2})$$

where, for notational convenience, the arguments  $\mathbf{q}$  and  $\omega$  of  $\underline{\mathbf{E}}$  and  $\underline{\mathbf{B}}$ , and the arguments  $-\mathbf{q}$  and  $\omega$  of  $\vec{G}_j$  and

$\vec{\Gamma}_j$  have been suppressed. Solving these linear relations for  $\underline{\mathbf{B}}(0) \times \mathbf{e}_z$  and  $\underline{\mathbf{B}}(d) \times \mathbf{e}_z$  in terms of  $\underline{\mathbf{E}}(0) \times \mathbf{e}_z$  and

$\underline{\mathbf{E}}(d) \times \mathbf{e}_z$ , we need only compare the result with Eq. (125) to verify Eqs. (143)–(146). This can be conveniently done by representing Eqs. (G1) and (G2) in the form

$$\mu^{-1} \begin{pmatrix} \underline{\mathbf{B}}(0) \times \mathbf{e}_z \\ \underline{\mathbf{B}}(d) \times \mathbf{e}_z \end{pmatrix}^T \cdot \begin{pmatrix} \vec{M} & \vec{N} \\ \vec{P} & \vec{S} \end{pmatrix} = \begin{pmatrix} \underline{\mathbf{E}}(0) \times \mathbf{e}_z \\ \underline{\mathbf{E}}(d) \times \mathbf{e}_z \end{pmatrix}^T \cdot \begin{pmatrix} \vec{T} & \vec{U} \\ \vec{V} & \vec{W} \end{pmatrix}, \quad (\text{G3})$$

where  $\vec{M}, \vec{N}, \vec{P}, \vec{S}, \vec{T}, \vec{U}, \vec{V}, \vec{W}$  are given in Eqs. (135)–(142). Rewriting Eq. (125) ( $z=0, d$ ) in an analogous form and applying to Eq. (G3) the inversion formula (F3), one

obtains

$$\begin{pmatrix} -\vec{Y}(0,0) & -\vec{Y}(0,d) \\ \vec{Y}(d,0) & \vec{Y}(d,d) \end{pmatrix} = \frac{1}{(2\pi)^2} \begin{pmatrix} \vec{T} & \vec{U} \\ \vec{V} & \vec{W} \end{pmatrix} \times \begin{pmatrix} \vec{P}^\# \cdot [\vec{M} \cdot \vec{P}^\# - \vec{N} \cdot \vec{S}^\#]^\# & \vec{M}^\# \cdot [\vec{P} \cdot \vec{M}^\# - \vec{S} \cdot \vec{N}^\#]^\# \\ \vec{S}^\# \cdot [\vec{N} \cdot \vec{S}^\# - \vec{M} \cdot \vec{P}^\#]^\# & \vec{N}^\# \cdot [\vec{S} \cdot \vec{N}^\# - \vec{P} \cdot \vec{M}^\#]^\# \end{pmatrix}, \quad (\text{G4})$$

which immediately leads to Eqs. (143)–(146).

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